

CASE PP/1-22181/US/A/RCE

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IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

IN RE APPLICATION OF

Group Art Unit: 1714

FRANÇOIS GUGUMUS

Examiner: T. Coo
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Confirmation No. 1010

APPLICATION NO: 09/823,041

FILED: MARCH 30, 2001

FOR: SYNERGISTIC MIXTURES OF UV-ABSORBERS

IN POLYOLEFINS

Commissioner for Patents

Mail Stop Duplicates

P.O. Box 1450

Alexandria, VA 22313-1450

DUPLICATE CLAIM OF PRIORITY UNDER 35 USC 119

Sir:

Applicants in the above-entitled application by their agent hereby claim priority under the International Convention of Europe (designating Germany) application No. 00810290.7, filed on April 4, 2000. This application is acknowledged in the Declaration of the instant case.

A certified copy of said European application is again submitted as per the Examiner's request. A certified copy was already submitted with payment of the Issue and Publication fees on July 19, 2004, however, it seems as though these papers were misplaced. We ask that this certified copy be acknowledged and notification be sent to us that it has been received by the U.S. Patent and Trademark Office.

Respectfully submitted,


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Bescheinigung

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Certificate

Attestation

Die angehefteten Unterlagen stimmen mit der ursprünglich eingereichten Fassung der auf dem nächsten Blatt bezeichneten europäischen Patentanmeldung überein.

The attached documents are exact copies of the European patent application described on the following page, as originally filed.

Les documents fixés à cette attestation sont conformes à la version initialement déposée de la demande de brevet européen spécifiée à la page suivante.

Patentanmeldung Nr. Patent application No. Demande de brevet n°

00810290.7

Der Präsident des Europäischen Patentamts;
Im Auftrag

For the President of the European Patent Office

Le Président de l'Office européen des brevets
p.o.

I.L.C. HATTEN-HECKMAN

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**Blatt 2 der Bescheinigung
Sheet 2 of the certificate
Page 2 de l'attestation**

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Anmeldung Nr.:
Application no.: **00810290.7**
Demande n°:

Anmeldetag:
Date of filing: **04/04/00**
Date de dépôt:

Anmelder:
Applicant(s):
Demandeur(s):
Ciba Specialty Chemicals Holding Inc.
4057 Basel
SWITZERLAND

Bezeichnung der Erfindung:
Title of the invention:
Titre de l'invention:
Synergistic mixtures of UV-absorbers in polyolefins

In Anspruch genommene Priorität(en) / Priority(ies) claimed / Priorité(s) revendiquée(s)

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Remarks:
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Synergistic Mixtures of UV-Absorbers in Polyolefins

The present invention relates to novel polyolefin compositions which contain mixtures of different UV absorbers.

The effects of atmospheric oxygen, moisture and, in particular, UV light result in degradation of the polymer material. This manifests itself, for example, in the loss of mechanical strength, changes in shade and finally total breakdown of the polymer article. As is known, it is possible to retard such degradation processes in polyolefins by the use of suitable stabilizers, and there are numerous prior art documents in this field.

In the coatings field, EP-A-0453 396 discloses that mixtures of hydroxyphenyl-benzotriazole with hydroxybenzophenone or with hydroxyphenyl-s-triazine UV-absorbers lead to synergistic mixtures which prevent the coatings life time unexpectedly long from degradation.

Although there are prior art documents, such as FR 2619 814, which generically disclose the combined use of oxalamide UV absorbers, particularly in coatings, with benzophenone or benzotriazole UV absorbers, there is however no suggestion in the prior art for the instant combinations in polyolefins.

It has now been found that certain combinations of UV absorbers of different chemical structures are capable of substantially preventing the degradation of polyolefins.

The combinations of the present invention provide an unexpected synergistic stabilization effect for polyolefin articles. The effect is not predictable from the absorption spectra and has not been observed with other UV absorber combinations in polyolefins.

One subject of the present invention is a polyolefin composition which comprises as UV absorber a mixture of

- a) at least one hydroxybenzophenone and at least one 2-hydroxyphenylbenzotriazole with the proviso that the polyolefin is a high density polyethylene or a polypropylene of the "Phillips" type or a polyethylene or polypropylene of the metallocene type and no polyvinylpyridin is present;

- b) at least one hydroxybenzophenone and at least one 2-hydroxyphenyltriazine, with the proviso that if the polyolefin is polypropylene, no polyvinylpyridin is present;
- c) at least one hydroxybenzophenone and at least one oxanilide;
- d) at least one 2-hydroxyphenylbenzotriazole and at least one oxanilide;
- e) at least one 2-hydroxyphenyltriazine and at least one oxanilide;
- f) at least one hydroxybenzophenone, at least one 2-hydroxyphenylbenzotriazole and at least one oxanilide;
- g) at least one hydroxybenzophenone, at least one 2-hydroxyphenylbenzotriazole and at least one 2-hydroxyphenyltriazine, with the proviso that if the polyolefin is polypropylene, no polyvinylpyridin is present;
- h) at least one hydroxybenzophenone, at least one oxanilide and at least one 2-hydroxyphenyltriazine; or
- i) at least one 2-hydroxyphenylbenzotriazole, at least one oxanilide and at least one 2-hydroxyphenyltriazine.

Suitable polyolefins are mentioned below.

1. Polymers of monoolefins and diolefins, for example polypropylene, polyisobutylene, polybut-1-ene, poly-4-methylpent-1-ene, polyisoprene or polybutadiene, as well as polymers of cycloolefins, for instance of cyclopentene or norbornene, polyethylene (which optionally can be crosslinked), for example high density polyethylene (HDPE), high density and high molecular weight polyethylene (HDPE-HMW), high density and ultrahigh molecular weight polyethylene (HDPE-UHMW), medium density polyethylene (MDPE), low density polyethylene (LDPE), linear low density polyethylene (LLDPE), (VLDPE) and (ULDPE), metallocene polyethylen (m-PE) in particular m-LLDPE and metallocene polypropylene (m-PP).

Polyolefins, i.e. the polymers of monoolefins exemplified in the preceding paragraph, preferably polyethylene and polypropylene, can be prepared by different, and especially by the following, methods:

- a) radical polymerisation (normally under high pressure and at elevated temperature).
- b) catalytic polymerisation using a catalyst that normally contains one or more than one metal of groups IVb, Vb, VIb or VIII of the Periodic Table. These metals usually have one or more than one ligand, typically oxides, halides, alcoholates, esters,

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ethers, amines, alkyls, alkenyls and/or aryls that may be either π - or σ -coordinated.

These metal complexes may be in the free form or fixed on substrates, typically on activated magnesium chloride, titanium(III) chloride, alumina or silicon oxide. These catalysts may be soluble or insoluble in the polymerisation medium. The catalysts can be used by themselves in the polymerisation or further activators may be used, typically metal alkyls, metal hydrides, metal alkyl halides, metal alkyl oxides or metal alkyloxanes, said metals being elements of groups Ia, IIa and/or IIIa of the Periodic Table. The activators may be modified conveniently with further ester, ether, amine or silyl ether groups. These catalyst systems are usually termed Phillips, Standard Oil Indiana, Ziegler (-Natta), TNZ (DuPont), metallocene or single site catalysts (SSC).

Preferred are Phillips and metallocene type polyolefins. In particular polyethylene and polypropylene prepared with these catalysts.

2. Mixtures of the polymers mentioned under 1), for example mixtures of polypropylene with polyisobutylene, polypropylene with polyethylene (for example PP/HDPE, PP/LDPE) and mixtures of different types of polyethylene (for example LDPE/HDPE), metallocene types with conventional types (for example m-PE/PE-LLD, m-PE/PE-LD, m-PP/conventional PP).

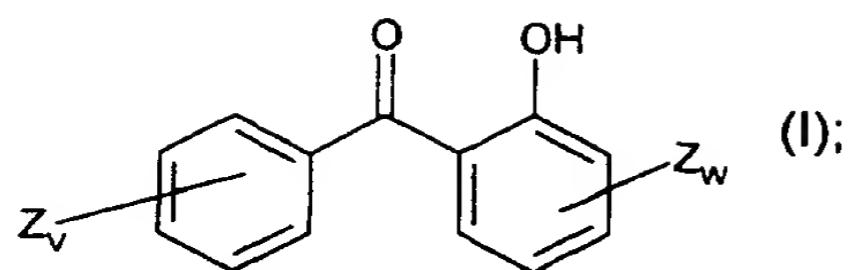
Preferably the polyolefin is polyethylene or polypropylene.

Preferred UV-absorber mixtures are:

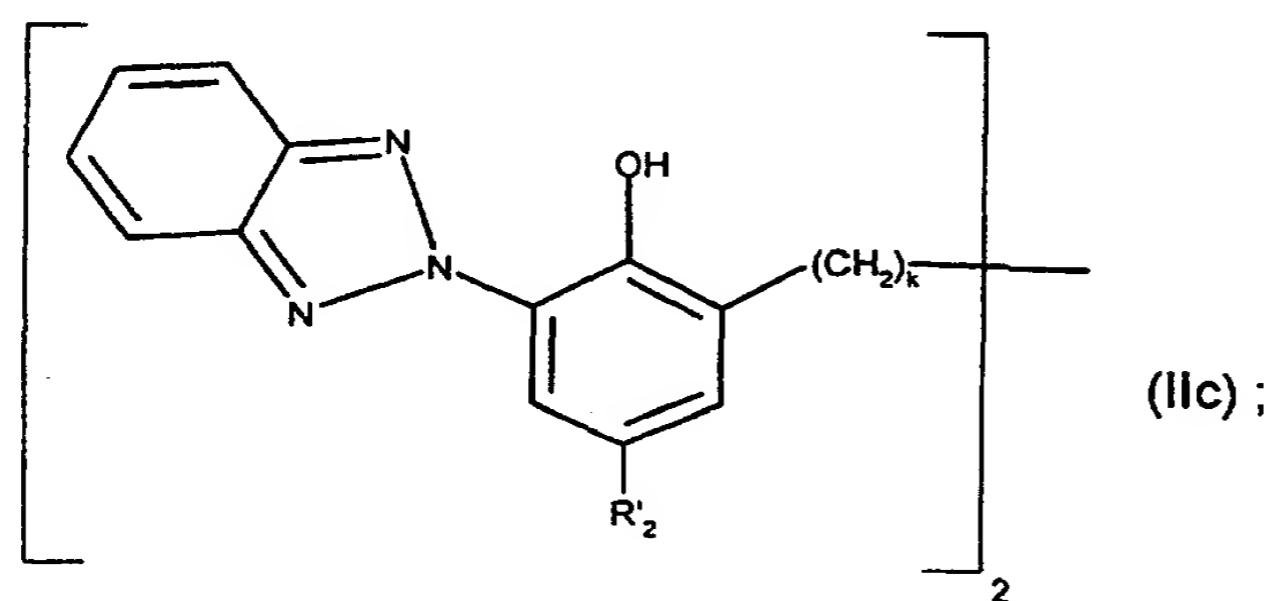
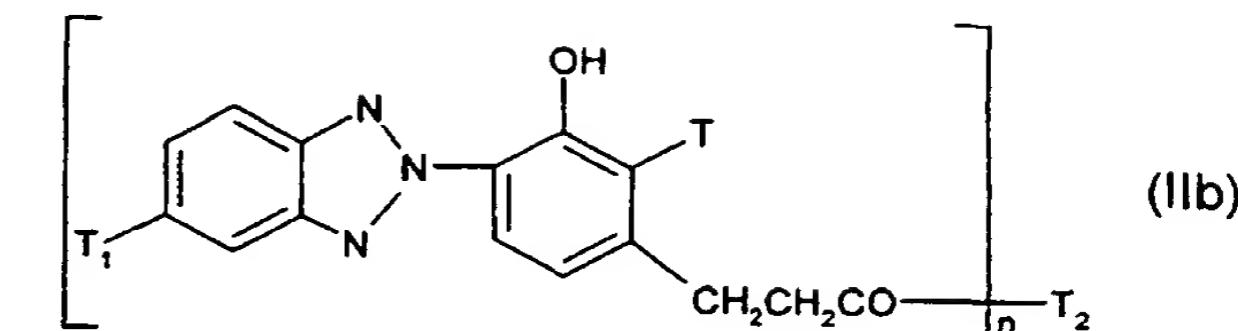
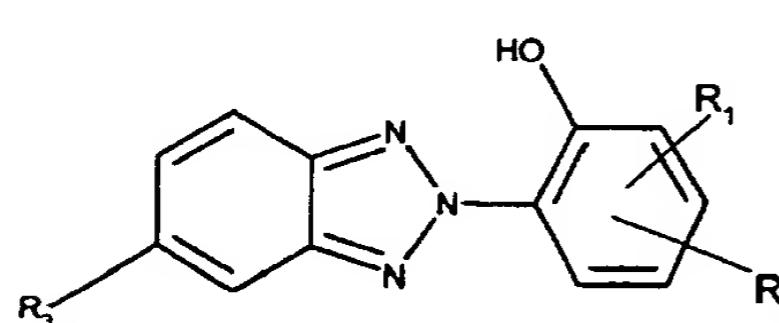
- a) at least one hydroxybenzophenone and at least one 2-hydroxyphenylbenzotriazole with the proviso that the polyolefin is a high density polyethylene or a polypropylene of the "Phillips" type or a metallocene polyethylene or polypropylene and no polyvinylpyridin is present;
- b) at least one hydroxybenzophenone and at least one 2-hydroxyphenyltriazine, with the proviso that if the polyolefin is polypropylene, no polyvinylpyridin is present and;
- e) at least one 2-hydroxyphenyltriazine and at least one oxanilide.

Preferred polyolefin compositions are wherein the hydroxybenzophenone is of formula I

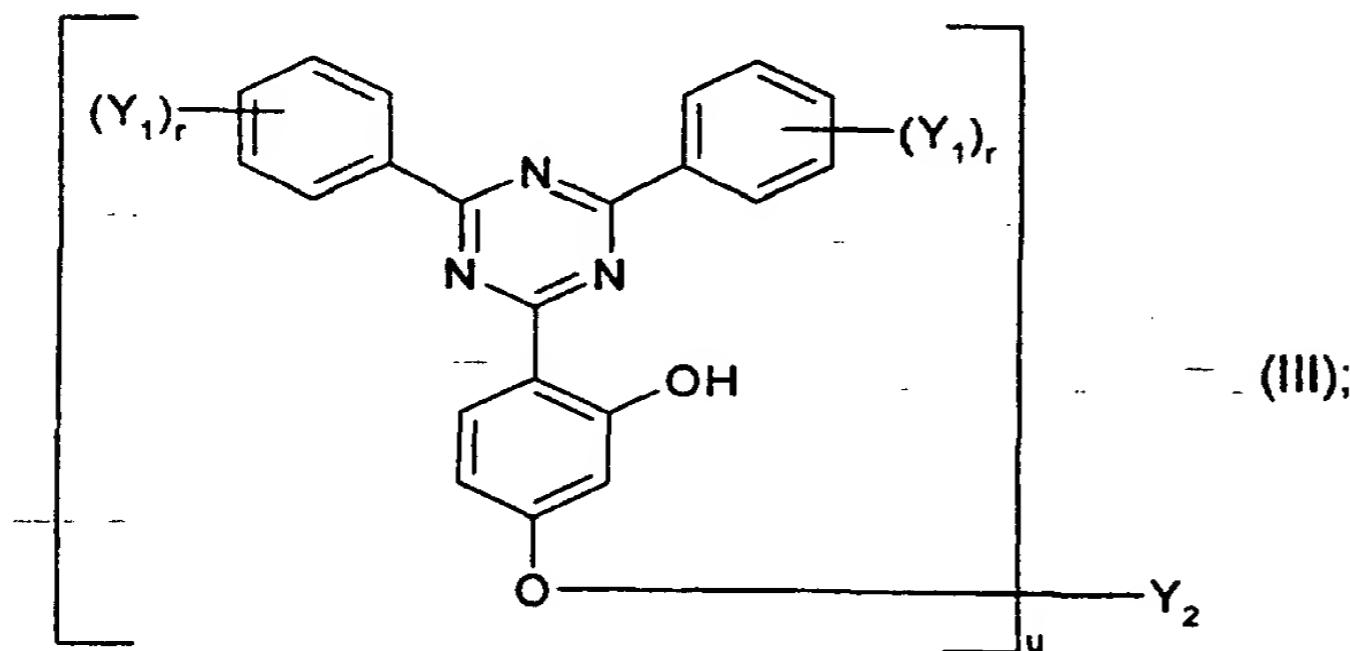
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the 2-hydroxyphenylbenzotriazole is of formula IIa, IIb or IIc

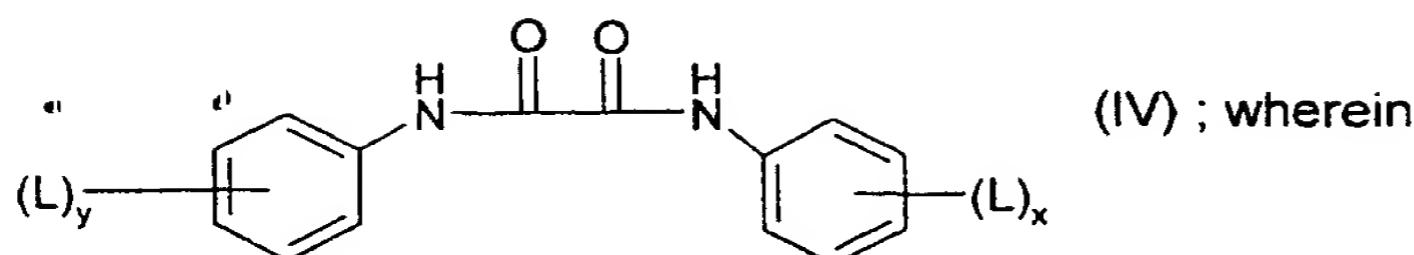


the 2-hydroxyphenyltriazine is of formula III



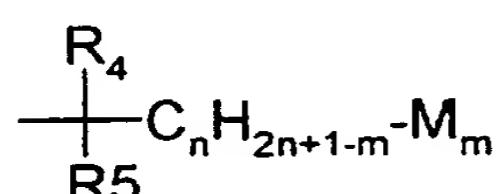
and the oxanilide is of formula (IV)

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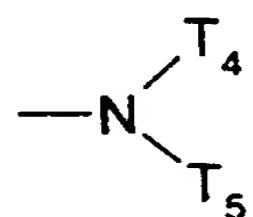
in the compounds of the formula (I) v is an integer from 1 to 3 and w is 1 or 2 and the substituents Z independently of one another are hydrogen, halogen, hydroxyl or alkoxy having 1 to 12 carbon atoms;

in the compounds of the formula (IIa), R₁ is hydrogen, alkyl having 1 to 24 carbon atoms, phenylalkyl having 1 to 4 carbon atoms in the alkyl moiety, cycloalkyl having 5 to 8 carbon atoms or a radical of the formula

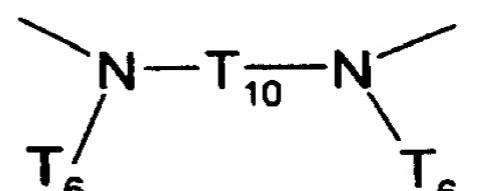


in which R₄ and R₅ independently of one another are alkyl having in each case 1 to 5 carbon atoms, or R₄, together with the radical C_nH_{2n+1-m}, forms a cycloalkyl radical having 5 to 12 carbon atoms, m is 1 or 2, n is an integer from 2 to 20 and M is a radical of the formula -COOR₆ in which R₆ is hydrogen, alkyl having 1 to 12 carbon atoms, alkoxyalkyl having in each case 1 to 20 carbon atoms in the alkyl moiety and in the alkoxy moiety or phenylalkyl having 1 to 4 carbon atoms in the alkyl moiety, R₂ is hydrogen, halogen, alkyl having 1 to 18 carbon atoms, and phenylalkyl having 1 to 4 carbon atoms in the alkyl moiety, and R₃ is hydrogen, chlorine, alkyl or alkoxy having in each case 1 to 4 carbon atoms or -COOR₆ in which R₆ is as defined above, at least one of the radicals R₁ and R₂ being other than hydrogen;

in the compounds of the formula (IIb) T is hydrogen or alkyl having 1 to 6 carbon atoms, T₁ is hydrogen, chlorine or alkyl or alkoxy having in each case 1 to 4 carbon atoms, n is 1 or 2 and, if n is 1, T₂ is chlorine or a radical of the formula -OT₃ or



and, if n is 2, T₂ is a radical of the formula

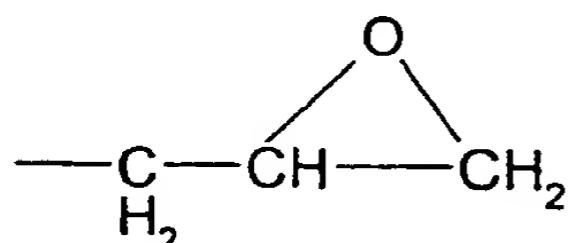


or -O-T₉-O-

in which T₃ is hydrogen, alkyl which has 1 to 18 carbon atoms and is unsubstituted or

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substituted by 1 to 3 hydroxyl groups or by $-OCOT_6$, alkyl which has 3 to 18 carbon atoms, is interrupted once or several times by $-O-$ or $-NT_6-$ and is unsubstituted or substituted by hydroxyl or $-OCOT_6$, cycloalkyl which has 5 to 12 carbon atoms and is unsubstituted or substituted by hydroxyl and/or alkyl having 1 to 4 carbon atoms, alkenyl which has 2 to 18 carbon atoms and is unsubstituted or substituted by hydroxyl, phenylalkyl having 1 to 4 carbon atoms in the alkyl moiety, or a radical of the formula $-CH_2CH(OH)-T_7$, or



, T_4 and T_5 independently of one another are hydrogen, alkyl

having 1 to 18 carbon atoms, alkyl which has 3 to 18 carbon atoms and is interrupted once or several times by $-O-$ or $-NT_6-$, cycloalkyl having 5 to 12 carbon atoms, phenyl, phenyl which is substituted by alkyl having 1 to 4 carbon atoms, alkenyl having 3 to 8 carbon atoms, phenylalkyl having 1 to 4 carbon atoms in the alkyl moiety or hydroxyalkyl having 2 to 4 carbon atoms, T_6 is hydrogen, alkyl having 1 to 18 carbon atoms, cycloalkyl having 5 to 12 carbon atoms, alkenyl having 3 to 8 carbon atoms, phenyl, phenyl which is substituted by alkyl having 1 to 4 carbon atoms, phenylalkyl having 1 to 4 carbon atoms in the alkyl moiety, T_7 is hydrogen, alkyl having 1 to 18 carbon atoms, phenyl which is unsubstituted or substituted by hydroxyl, phenylalkyl having 1 to 4 carbon atoms in the alkyl moiety, or $-CH_2OT_8$, T_8 is alkyl having 1 to 18 carbon atoms, alkenyl having 3 to 8 carbon atoms, cycloalkyl having 5 to 10 carbon atoms, phenyl, phenyl which is substituted by alkyl having 1 to 4 carbon atoms, or phenylalkyl having 1 to 4 carbon atoms in the alkyl moiety, T_9 is alkylene having 2 to 8 carbon atoms, alkenylene having 4 to 8 carbon atoms, alkynylene having 4 carbon atoms, cyclohexylene, alkylene which has 2 to 8 carbon atoms and is interrupted once or several times by $-O-$, or a radical of the formula $-CH_2CH(OH)CH_2OT_{11}OCH_2CH(OH)CH_2-$ or $-CH_2-C(CH_2OH)_2-CH_2-$, T_{10} is alkylene which has 2 to 20 carbon atoms and can be interrupted once or several times by $-O-$, or cyclohexylene, T_{11} is alkylene having 2 to 8 carbon atoms, alkylene which has 2 to 18 carbon atoms and is interrupted once or several times by $-O-$, 1,3-cyclohexylene, 1,4-cyclohexylene, 1,3-phenylene or 1,4-phenylene, or T_{10} and T_6 , together with the two nitrogen atoms, are a piperazine ring;

in the compounds of formula (IIc) R'_2 is C_1-C_{12} alkyl and k is a number from 1 to 4;

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in the compounds of the formula (III) u is 1 or 2 and r is an integer from 1 to 3, the substituents Y₁ independently of one another are hydrogen, hydroxyl, halogenomethyl, alkyl having 1 to 12 carbon atoms, alkoxy having 1 to 18 carbon atoms, or halogen, if u is 1, Y₂ is alkyl having 1 to 18 carbon atoms, phenoxy which is unsubstituted or substituted by hydroxyl, alkoxy having 1 to 18 carbon atoms, or halogen, or is substituted by alkyl or alkoxy having in each case 1 to 18 carbon atoms or halogen, alkyl which has 1 to 12 carbon atoms and is substituted by -COOH, -COOY₈, -CONH₂, -CONHY₉, -CONY₉Y₁₀, -NH₂, -NHY₉, -NY₉Y₁₀, -NHCOY₁₁, -CN and/or -OCOY₁₁, alkyl which has 4 to 20 carbon atoms, is interrupted by one or more oxygen atoms and is unsubstituted or substituted by hydroxyl or alkoxy having 1 to 12 carbon atoms, alkenyl having 3 to 6 carbon atoms, glycidyl, cyclohexyl which is unsubstituted or substituted by hydroxyl, alkyl having 1 to 4 carbon atoms and/or -OCOY₁₁, phenylalkyl which has 1 to 5 carbon atoms in the alkyl moiety and is unsubstituted or substituted by hydroxyl, chlorine and/or methyl, -COY₁₂ or -SO₂Y₁₃, or, if u is 2, Y₂ is alkylene having 2 to 16 carbon atoms, alkenylene having 4 to 12 carbon atoms, xylylene, alkylene which has 3 to 20 carbon atoms, is interrupted by one or more -O- atoms and/or is substituted by hydroxyl, -CH₂CH(OH)CH₂-O-Y₁₅-OCH₂CH(OH)CH₂, -CO-Y₁₆-CO-, -CO-NH-Y₁₇-NH-CO- or -(CH₂)_m-CO₂-Y₁₈-OCO-(CH₂)_m, in which m is 1, 2 or 3, Y₈ is alkyl having 1 to 18 carbon atoms, alkenyl having 3 to 18 carbon atoms, alkyl which has 3 to 20 carbon atoms, is interrupted by one or more oxygen or sulfur atoms or -NT₆- and/or is substituted by hydroxyl, alkyl which has 1 to 4 carbon atoms and is substituted by -P(O)(OY₁₄)₂, -NY₉Y₁₀ or -OCOY₁₁ and/or hydroxyl, alkenyl having 3 to 18 carbon atoms, glycidyl, or phenylalkyl having 1 to 5 carbon atoms in the alkyl moiety, Y₉ and Y₁₀ independently of one another are alkyl having 1 to 12 carbon atoms, alkoxyalkyl having 3 to 12 carbon atoms, dialkylaminoalkyl having 4 to 16 carbon atoms or cyclohexyl having 5 to 12 carbon atoms, or Y₉ and Y₁₀ together are alkylene, oxaalkylene or azaalkylene having in each case 3 to 9 carbon atoms, Y₁₁ is alkyl having 1 to 18 carbon atoms, alkenyl having 2 to 18 carbon atoms or phenyl, Y₁₂ is alkyl having 1 to 18 carbon atoms, alkenyl having 2 to 18 carbon atoms, phenyl, alkoxy having 1 to 12 carbon atoms, phenoxy, alkylamino having 1 to 12 carbon atoms or phenylamino, Y₁₃ is alkyl having 1 to 18 carbon atoms, phenyl or alkylphenyl having 1 to 8 carbon atoms in the alkyl radical, Y₁₄ is alkyl having 1 to 12 carbon atoms or phenyl, Y₁₅ is alkylene having 2 to 10 carbon atoms, phenylene or a group -phenylene-M-phenylene- in which M is -O-, -S-, -SO₂-, -CH₂- or -C(CH₃)₂-, Y₁₆ is alkylene, oxaalkylene or thiaalkylene having in each case 2 to 10 carbon atoms, phenylene or alkenylene having 2 to 6 carbon atoms, Y₁₇ is alkylene having 2 to 10 carbon atoms, phenylene or alkylphenylene having 1 to

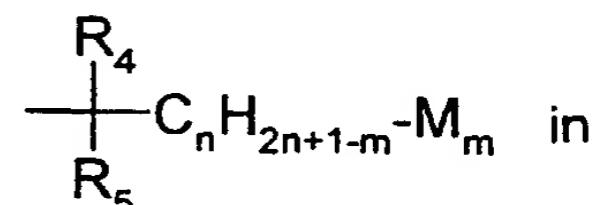
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11 carbon atoms in the alkyl moiety, and Y₁₈ is alkylene having 2 to 10 carbon atoms or alkylene which has 4 to 20 carbon atoms and is interrupted once or several times by oxygen;

in the compounds of the formula (IV) x is an integer from 1 to 3 and the substituents L independently of one another are hydrogen, alkyl, alkoxy or alkylthio having in each case 1 to 22 carbon atoms, phenoxy or phenylthio.

In the compounds of the formula (IIa) R₁ can be hydrogen or alkyl having 1 to 24 carbon atoms, such as methyl, ethyl, propyl, butyl, hexyl, octyl, nonyl, dodecyl, tetradecyl, hexadecyl, octadecyl, nonadecyl and eicosyl and also corresponding branched isomers. Furthermore, in addition to phenylalkyl having 1 to 4 carbon atoms in the alkyl moiety, for example benzyl, R₁ can also be cycloalkyl having 5 to 8 carbon atoms, for example

cyclopentyl, cyclohexyl and cyclooctyl, or a radical of the formula



which R₄ and R₅ independently of one another are alkyl having in each case 1 to 5 carbon atoms, in particular methyl, or R₄, together with the C_nH_{2n+1-m} radical, forms a cycloalkyl radical having 5 to 12 carbon atoms, for example cyclohexyl, cyclooctyl and cyclodecyl. M is a radical of the formula -COOR₆ in which R₆ is not only hydrogen but also alkyl having 1 to 12 carbon atoms or alkoxyalkyl having 1 to 20 carbon atoms in each of the alkyl and alkoxy moieties. Suitable alkyl radicals R₆ are those enumerated for R₁. Examples of suitable alkoxyalkyl groups are -C₂H₄OC₂H₅, -C₂H₄OC₈H₁₇ and -C₄H₈OC₄H₉. As phenylalkyl having 1 to 4 carbon atoms, R₆ is, for example, benzyl, cumyl, α-methylbenzyl or phenylbutyl.

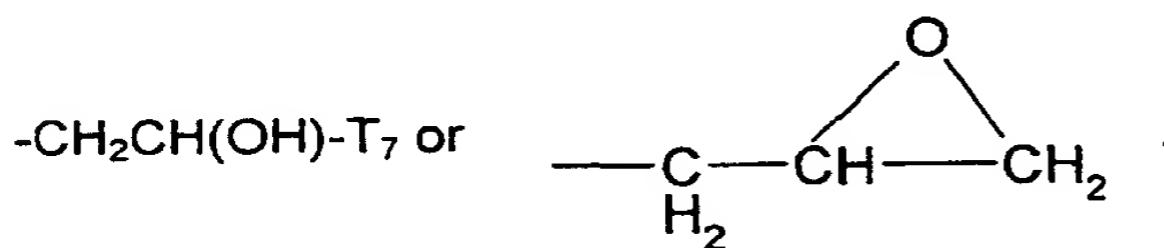
In addition to hydrogen and halogen, for example chlorine and bromine, R₂ can also be alkyl having 1 to 18 carbon atoms. Examples of such alkyl radicals are indicated in the definitions of R₁. R₂ can also be phenylalkyl having 1 to 4 carbon atoms in the alkyl moiety, for example benzyl, α-methylbenzyl and cumyl.

At least one of the radicals R₁ and R₂ must be other than hydrogen.

In addition to hydrogen or chlorine, R₃ is also alkyl or alkoxy having in each case 1 to 4 carbon atoms, for example methyl, butyl, methoxy and ethoxy, and also -COOR₆.

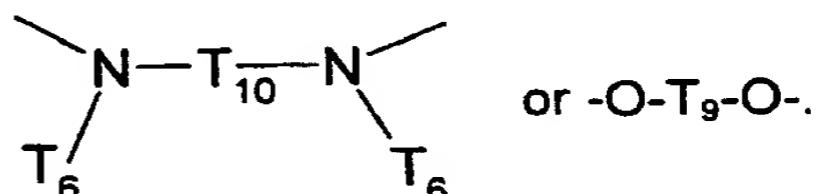
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In the compounds of the formula (IIb) T is hydrogen or alkyl having 1 to 6 carbon atoms, such as methyl and butyl, T₁ is not only hydrogen or chlorine, but also alkyl or alkoxy having in each case 1 to 4 carbon atoms, for example methyl, methoxy and butoxy, and, if n is 1, T₂ is chlorine or a radical of the formula -OT₃ or -NT₄T₅. T₃ is here hydrogen or alkyl having 1 to 18 carbon atoms (cf. the definition of R₁). These alkyl radicals can be substituted by 1 to 3 hydroxyl groups or by a radical -OCOT₆. Furthermore, T₃ can be alkyl having 3 to 18 carbon atoms (cf. the definition of R₁) which is interrupted once or several times by -O- or -NT₆- and is unsubstituted or substituted by hydroxyl or -OCOT₆. Examples of T₃ as cycloalkyl are cyclopentyl, cyclohexyl or cyclooctyl. T₃ can also be alkenyl having 2 to 18 carbon atoms. Suitable alkenyl radicals are derived from the alkyl radicals enumerated in the definitions of R₁. These alkenyl radicals can be substituted by hydroxyl. Examples of T₃ as phenylalkyl are benzyl, phenylethyl, cumyl, α-methylbenzyl or benzyl. T₃ can also be a radical of the formula



Like T₃, T₄ and T₅ can, independently of one another, be not only hydrogen but also alkyl having 1 to 18 carbon atoms or alkyl which has 3 to 18 carbon atoms and is interrupted once or several times by -O- or -NT₆- . T₄ and T₅ can also be cycloalkyl having 5 to 12 carbon atoms, for example cyclopentyl, cyclohexyl and cyclooctyl. Examples of T₄ and T₅ as alkenyl groups can be found in the illustrations of T₃. Examples of T₄ and T₅ as phenylalkyl having 1 to 4 carbon atoms in the alkyl moiety are benzyl or phenylbutyl. Finally, these substituents can also be hydroxyalkyl having 1 to 3 carbon atoms.

If n is 2, T₂ is a divalent radical of the formula



In addition to hydrogen, T₆ (see above also) is alkyl, cycloalkyl, alkenyl, aryl or phenylalkyl; examples of such radicals have already been given above.

In addition to hydrogen and the phenylalkyl radicals and long-chain alkyl radicals mentioned above, T₇ can be phenyl or hydroxyphenyl and also -CH₂OT₈ in which T₈ can be one of the

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alkyl, alkenyl, cycloalkyl, aryl or phenylalkyl radicals enumerated.

The divalent radical T_9 can be alkylene having 2 to 8 carbon atoms, and such radicals can also be branched. This also applies to the alkenylene and alkynylene radicals T_9 . As well as cyclohexylene, T_9 can also be a radical of the formula -



T_{10} is a divalent radical and, in addition to cyclohexylene, is also alkylene which has 2 to 20 carbon atoms and which can be interrupted once or several times by $-\text{O}-$. Suitable alkylene radicals are derived from the alkyl radicals mentioned in the definitions of R_1 .

T_{11} is also an alkylene radical. It contains 2 to 8 carbon atoms or, if it is interrupted once or several times by $-\text{O}-$, 4 to 10 carbon atoms. T_{11} is also 1,3-cyclohexylene, 1,4-cyclohexylene, 1,3-phenylene or 1,4-phenylene.

Together with the two nitrogen atoms, T_6 and T_{10} can also be a piperazine ring.

Examples of alkyl, alkoxy, phenylalkyl, alkylene, alkenylene, alkoxyalkyl and cycloalkyl radicals and also alkylthio, oxaalkylene or azoalkylene radicals in the compounds of the formulae (2), (3) and (4) can be deduced from the above statements.

The UV absorbers of the formulae (I), (IIa), (IIb), (III), and (IV) are known per se and are described, together with their preparation, in, for example, EP-A-323 408, EP-A-57 160, US Patent Application Serial No. 07/446 369 (prior.: 5.12.89), US-A 4 619 956, DE-A 31 35 810 and GB-A 1 336 391. Preferred meanings of substituents and individual compounds can be deduced from the documents mentioned. They can be incorporated into polyolefins by customary methods which are known per se. The preparation may be carried out in a manner known per se by mixing the said components and, if desired, further additives with the polymer using devices known per se, such as calenders, mixers, kneaders, extruders and the like. The additives may be added individually or in admixture with one another. It is also possible to use so-called master batches.

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In certain cases it can be advantageous to use even a mixture of compounds of all 4 classes of UV-absorbers (hydroxybenzophenone, 2-hydroxyphenyltriazine, oxanilide and 2-hydroxyphenylbenzotriazole).

The polyolefin composition obtainable according to the present invention can be made into the desired form in known manner. Such processes include, for example, grinding, calendering, extruding, injection-moulding, sintering, compression/sintering or spinning, also extrusion blow-moulding.

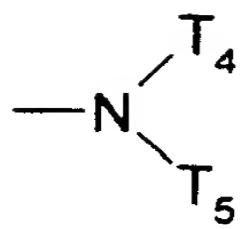
The amounts of UV absorber to be selected can depend on the nature of the polyolefin composition and the requirements for its stability. The individual UV absorber components can be added on their own or as a mixture to the appropriate polyolefin compositions. The customary other additives, such as processing stabilizers, antioxidants and other light stabilisers, can also be added to the polyolefin compositions, without thereby impairing the protective action of the UV absorber mixtures employed in accordance with the invention.

Preferred is a polyolefin composition wherein the substituent R₁ or R₂ in the compounds of the formula (IIa) is located in the ortho-position or para-position relative to the hydroxyl group.

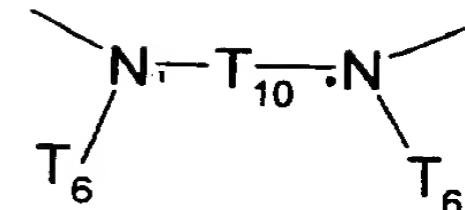
Also preferred is a polyolefin composition, in which, in the compounds of the formula (IIa), R₁ is hydrogen or alkyl having 1 to 20 carbon atoms, R₂ is hydrogen, alkyl having 1 to 18 carbon atoms or phenylalkyl having 1 to 4 carbon atoms in the alkyl moiety and R₃ is hydrogen, chlorine or alkyl having 1 to 4 carbon atoms. Preference is given to compounds, in which R₁ is in the ortho-position relative to the hydroxyl group and is hydrogen or alkyl having 4 to 12 carbon atoms, R₂ is in the para-position relative to the hydroxyl group and is alkyl having 1 to 6 carbon atoms or cumyl and R₃ is hydrogen or chlorine. More preferably R₁ is alkyl having 8 to 12 carbon atoms, R₂ is alkyl having 4 to 6 carbon atoms and R₃ is hydrogen.

Other preferred polyolefin compositions are those, in which, in the compounds of the formula (IIb), T is alkyl having 1 to 6 carbon atoms, T₁ is hydrogen, chlorine or alkyl having 1 to 4 carbon atoms, n is 1 or 2 and, if n is 1, T₂ is one of the radicals of the formula -OT₃ or

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and, if n is 2, T₂ is a radical of the formula -O-T₉-O- or



in which T₃ is hydrogen, alkyl having 1 to 18 carbon atoms or alkyl which has 3 to 18 carbon atoms and is interrupted once or several times by -O-, T₄ and T₅ independently of one another are hydrogen, alkyl having 1 to 18 carbon atoms, alkenyl having 3 to 8 carbon atoms or hydroxyalkyl having 2 to 4 carbon atoms, T₆ is hydrogen or alkyl having 1 to 6 carbon atoms, and T₉ and T₁₀ are alkylene having 2 to 8 carbon atoms, alkenylene having 4 to 8 carbon atoms or alkylene which has 2 to 18 carbon atoms and is interrupted once or several times by -O-..

More preferably, in which T is alkyl having 1 to 4 carbon atoms, T₁ is hydrogen or chlorine and T₂ is one of the radicals of the formula -OT₃ or -O-T₉-O- in which T₃ is hydrogen, alkyl having 1 to 12 carbon atoms or a radical of the formula -(OC₂H₄)₄₋₈-H and T₉ is alkylene having 2 to 8 carbon atoms.

Most preferably T₂ is a radical of the formula -OT₃ and T₃ is alkyl having 1 to 12 carbon atoms or a radical of the formula -(OC₂H₄)₄₋₈-H.

Further preferred polyolefin compositions are those, in which, in the compounds of the formula (III), the substituents Y₁ are hydrogen, alkyl having 1 to 12 carbon atoms or halogen, if u is 1, Y₂ is alkyl having 1 to 18 carbon atoms, alkyl which has 1 to 12 carbon atoms and is substituted by hydroxyl, alkoxy having 1 to 18 carbon atoms, -COOY₈, -CONY₉Y₁₀ and/or -OCOY₁₁, glycidyl or phenylalkyl having 1 to 4 carbon atoms in the alkyl moiety, or, if u is 2, Y₂ is alkylene having 2 to 16 carbon atoms, alkenylene having 4 to 12 carbon atoms, xylylene or alkylene which has 3 to 20 carbon atoms, is interrupted by one or more -O- atoms and/or is substituted by hydroxyl, the substituents Y₈ to Y₁₁ being as defined above.

Preferably Y₁ are hydrogen, alkyl having 1 to 4 carbon atoms or chlorine, if u is 1, Y₂ is alkyl which has 1 to 4 carbon atoms and is unsubstituted or substituted by hydroxyl, alkoxy having 1 to 18 carbon atoms, -COOY₈, -CONY₉Y₁₀ and/or -OCOY₁₁, glycidyl or benzyl, or, if u is 2, Y₂ is alkylene having 6 to 12 carbon atoms, butenylene, xylylene or alkylene which has 3 to 20 carbon atoms, is interrupted by one or more -O- atoms and/or is substituted by hydroxyl, Y₈ being alkyl having 4 to 12 carbon atoms, alkenylene having 12 to 18 carbon atoms or alkyl which has 6 to 20 carbon atoms, is interrupted by one or more -O- atoms

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and/or is substituted by hydroxyl, Y_9 and Y_{10} independently of one another being alkyl having 4 to 8 carbon atoms and Y_{11} being alkyl having 1 to 8 carbon atoms or alkenyl having 2 or 3 carbon atoms.

More preferably u is 1 and r is 2, Y_1 is alkyl having 1 to 4 carbon atoms and Y_2 is alkyl having 1 to 18 carbon atoms or alkyl which has 1 to 12 carbon atoms and is substituted by hydroxyl, alkoxy having 1 to 18 carbon atoms, $-COOY_8$ and/or $-OCOY_{11}$, Y_8 being alkyl having 1 to 18 carbon atoms, alkenyl having 3 to 18 carbon atoms or alkyl which has 3 to 20 carbon atoms, is interrupted by one or more oxygen atoms and/or is substituted by hydroxyl, and Y_{11} being alkenyl having 2 to 18 carbon atoms.

Particularly preferred are the compounds of formula (III), in which Y_2 is alkyl having 1 to 8 carbon atoms or alkyl which has 1 to 4 carbon atoms and is substituted by hydroxyl, alkoxy having 12 to 15 carbon atoms, $-COOY_8$ and/or $-OCOY_{11}$, Y_8 being alkyl having 8 to 12 carbon atoms, alkenyl having 12 to 18 carbon atoms or alkyl which has 5 to 10 carbon atoms, is interrupted by an oxygen atom and is substituted by hydroxyl, and Y_{11} being alkenyl having 2 to 8 carbon atoms.

Most preferred are compounds of formula (III), in which Y_1 is methyl and Y_2 is an octyl radical or alkyl which has 1 to 3 carbon atoms and is substituted by hydroxyl, alkoxy having 13 or 15 carbon atoms, $-COOY_8$ and/or $-OCOY_{11}$, Y_8 being a decyl or octadecenyl radical or alkyl which has 7 carbon atoms and is substituted by hydroxyl and interrupted by an oxygen atom, and Y_{11} being propenyl.

A further preferred polyolefin composition is, in which, in the compounds of the formula (I), v and w independently of one another are 1 or 2 and the substituents Z independently of one another are hydrogen, halogen or alkoxy having 1 to 12 carbon atoms.

Also preferred is a composition, in which, in the compounds of the formula (IV), x and y are 1 or 2 and the substituents L independently of one another are hydrogen or alkyl having in each case 1 to 12 carbon atoms.

Individual UV-absorbers suitable in the present invention are given below.

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2-Hydroxybenzophenones, for example the 4-hydroxy, 4-methoxy, 4-octyloxy, 4-decyloxy, 4-dodecyloxy, 4-benzyloxy, 4,2',4'-trihydroxy and 2'-hydroxy-4,4'-dimethoxy derivatives.

2-(2'-Hydroxyphenyl)benzotriazoles, for example 2-(2'-hydroxy-5'-methylphenyl)-benzotriazole, 2-(3',5'-di-tert-butyl-2'-hydroxyphenyl)benzotriazole, 2-(5'-tert-butyl-2'-hydroxyphenyl)benzotriazole, 2-(2'-hydroxy-5'-(1,1,3,3-tetramethylbutyl)phenyl)benzotriazole, 2-(3',5'-di-tert-butyl-2'-hydroxyphenyl)-5-chloro-benzotriazole, 2-(3'-tert-butyl-2'-hydroxy-5'-methylphenyl)-5-chloro-benzotriazole, 2-(3'-sec-butyl-5'-tert-butyl-2'-hydroxyphenyl)benzotriazole, 2-(2'-hydroxy-4'-octyloxyphenyl)benzotriazole, 2-(3',5'-di-tert-amyl-2'-hydroxyphenyl)benzotriazole, 2-(3',5'-bis-(α,α -dimethylbenzyl)-2'-hydroxyphenyl)benzotriazole, 2-(3'-tert-butyl-2'-hydroxy-5'-(2-octyloxycarbonylethyl)phenyl)-5-chloro-benzotriazole, 2-(3'-tert-butyl-5'-[2-(2-ethylhexyloxy)-carbonylethyl]-2'-hydroxyphenyl)-5-chloro-benzotriazole, 2-(3'-tert-butyl-2'-hydroxy-5'-(2-methoxycarbonylethyl)phenyl)-5-chloro-benzotriazole, 2-(3'-tert-butyl-2'-hydroxy-5'-(2-methoxycarbonylethyl)phenyl)benzotriazole, 2-(3'-tert-butyl-2'-hydroxy-5'-(2-octyloxycarbonylethyl)phenyl)benzotriazole, 2-(3'-tert-butyl-5'-[2-(2-ethylhexyloxy)carbonylethyl]-2'-hydroxyphenyl)benzotriazole, 2-(3'-dodecyl-2'-hydroxy-5'-methylphenyl)benzotriazole, 2-(3'-tert-butyl-2'-hydroxy-5'-(2-isooctyloxycarbonylethyl)phenyl)benzotriazole, 2,2'-methyleno-bis-[4-(1,1,3,3-tetramethylbutyl)-6-benzotriazole-2-ylphenol]; the transesterification product of 2-[3'-tert-butyl-5'-(2-methoxycarbonylethyl)-2'-hydroxyphenyl]-2H-benzotriazole with polyethylene glycol 300; $[R-\text{CH}_2\text{CH}_2-\text{COO}-\text{CH}_2\text{CH}_2]_2$ where R = 3'-tert-butyl-4'-hydroxy-5'-2H-benzotriazol-2-ylphenyl, 2-[2'-hydroxy-3'-(α,α -dimethylbenzyl)-5'-(1,1,3,3-tetramethylbutyl)-phenyl]benzotriazole; 2-[2'-hydroxy-3'-(1,1,3,3-tetramethylbutyl)-5'-(α,α -dimethylbenzyl)-phenyl]benzotriazole.

2-(2-Hydroxyphenyl)-1,3,5-triazines, for example 2,4,6-tris(2-hydroxy-4-octyloxyphenyl)-1,3,5-triazine, 2-(2-hydroxy-4-octyloxyphenyl)-4,6-bis(2,4-dimethylphenyl)-1,3,5-triazine, 2-(2,4-dihydroxyphenyl)-4,6-bis(2,4-dimethylphenyl)-1,3,5-triazine, 2,4-bis(2-hydroxy-4-propoxyphenyl)-6-(2,4-dimethylphenyl)-1,3,5-triazine, 2-(2-hydroxy-4-octyloxyphenyl)-4,6-bis(4-methylphenyl)-1,3,5-triazine, 2-(2-hydroxy-4-dodecyloxyphenyl)-4,6-bis(2,4-dimethylphenyl)-1,3,5-triazine, 2-(2-hydroxy-4-tridecyloxyphenyl)-4,6-bis(2,4-dimethylphenyl)-1,3,5-triazine, 2-[2-hydroxy-4-(2-hydroxy-3-butyloxy-propoxy)phenyl]-4,6-bis(2,4-dimethyl)-1,3,5-triazine, 2-[2-hydroxy-4-(2-hydroxy-3-octyloxy-propoxy)phenyl]-4,6-bis(2,4-dimethyl)-1,3,5-triazine, 2-[4-(dodecyloxy/tridecyloxy-2-hydroxypropoxy)-2-hydroxy-phenyl]-4,6-bis(2,4-di-

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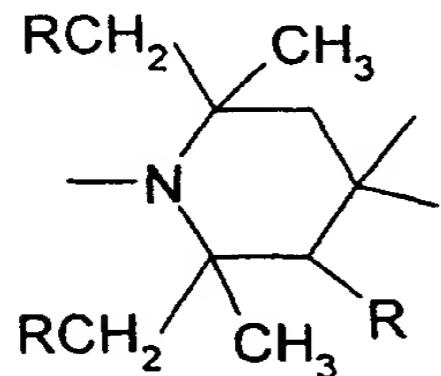
methylphenyl)-1,3,5-triazine, 2-[2-hydroxy-4-(2-hydroxy-3-dodecyloxy-propoxy)phenyl]-4,6-bis(2,4-dimethylphenyl)-1,3,5-triazine, 2-(2-hydroxy-4-hexyloxy)phenyl-4,6-diphenyl-1,3,5-triazine, 2-(2-hydroxy-4-methoxyphenyl)-4,6-diphenyl-1,3,5-triazine, 2,4,6-tris[2-hydroxy-4-(3-butoxy-2-hydroxy-propoxy)phenyl]-1,3,5-triazine, 2-(2-hydroxyphenyl)-4-(4-methoxyphenyl)-6-phenyl-1,3,5-triazine, 2-{2-hydroxy-4-[3-(2-ethylhexyl-1-oxy)-2-hydroxypropyloxy]phenyl}-4,6-bis(2,4-dimethylphenyl)-1,3,5-triazine.

Oxamides, for example 4,4'-dioctyloxyoxanilide, 2,2'-diethoxyoxanilide, 2,2'-dioctyloxy-5,5'-di-tert-butoxanilide, 2,2'-didodecyloxy-5,5'-di-tert-butoxanilide, 2-ethoxy-2'-ethyloxanilide, N,N'-bis(3-dimethylaminopropyl)oxamide, 2-ethoxy-5-tert-butyl-2'-ethoxanilide and its mixture with 2-ethoxy-2'-ethyl-5,4'-di-tert-butoxanilide, mixtures of o- and p-methoxy-disubstituted oxanilides and mixtures of o- and p-ethoxy-disubstituted oxanilides.

Preferably the amount of the individual UV absorber in the mixture is from 20% to 80% based on the weight of the mixture, with the proviso that the sum of the individual components adds to 100%.

Preferably the total amount of the UV-absorber mixture is from 0.005 to 5% based on the weight of the polymer.

Also preferred is a polyolefin composition, which additionally contains at least one sterically hindered amine, in particular an amine of this type containing at least one radical of the formula



in which R is hydrogen or methyl.

Examples of such sterically hindered amines are bis(2,2,6,6-tetramethyl-4-piperidyl)-sebacate, bis(2,2,6,6-tetramethyl-4-piperidyl)succinate, bis(1,2,2,6,6-pentamethyl-4-piperidyl)sebacate, bis(1-octyloxy-2,2,6,6-tetramethyl-4-piperidyl)sebacate, bis(1,2,2,6,6-pentamethyl-4-piperidyl) n-butyl-3,5-di-tert-butyl-4-hydroxybenzylmalonate, the condensate of 1-(2-hydroxyethyl)-2,2,6,6-tetramethyl-4-hydroxypiperidine and succinic acid, linear or

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cyclic condensates of N,N'-bis(2,2,6,6-tetramethyl-4-piperidyl)hexamethylenediamine and 4-tert-octylamino-2,6-dichloro-1,3,5-triazine, tris(2,2,6,6-tetramethyl-4-piperidyl)nitrioltriacetate, tetrakis(2,2,6,6-tetramethyl-4-piperidyl)-1,2,3,4-butane-tetracarboxylate, 1,1'-(1,2-ethanediyl)-bis(3,3,5,5-tetramethylpiperazinone), 4-benzoyl-2,2,6,6-tetramethylpiperidine, 4-stearylxy-2,2,6,6-tetramethylpiperidine, bis(1,2,2,6,6-pentamethylpiperidyl)-2-n-butyl-2-(2-hydroxy-3,5-di-tert-butylbenzyl)malonate, 3-n-octyl-7,7,9,9-tetramethyl-1,3,8-triazaspiro[4.5]-decan-2,4-dione, bis(1-octyloxy-2,2,6,6-tetramethylpiperidyl)sebacate, bis(1-octyloxy-2,2,6,6-tetramethylpiperidyl)succinate, linear or cyclic condensates of N,N'-bis-(2,2,6,6-tetramethyl-4-piperidyl)hexamethylenediamine and 4-morpholino-2,6-dichloro-1,3,5-triazine, the condensate of 2-chloro-4,6-bis(4-n-butylamino-2,2,6,6-tetramethylpiperidyl)-1,3,5-triazine and 1,2-bis(3-aminopropylamino)ethane, the condensate of 2-chloro-4,6-di-(4-n-butylamino-1,2,2,6,6-pentamethylpiperidyl)-1,3,5-triazine and 1,2-bis-(3-aminopropylamino)ethane, 8-acetyl-3-dodecyl-7,7,9,9-tetramethyl-1,3,8-triazaspiro[4.5]decane-2,4-dione, 3-dodecyl-1-(2,2,6,6-tetramethyl-4-piperidyl)pyrrolidin-2,5-dione, 3-dodecyl-1-(1,2,2,6,6-pentamethyl-4-piperidyl)pyrrolidine-2,5-dione, a mixture of 4-hexadecyloxy- and 4-stearylxy-2,2,6,6-tetramethylpiperidine, a condensation product of N,N'-bis(2,2,6,6-tetramethyl-4-piperidyl)hexamethylenediamine and 4-cyclohexylamino-2,6-dichloro-1,3,5-triazine, a condensation product of 1,2-bis(3-aminopropylamino)ethane and 2,4,6-trichloro-1,3,5-triazine as well as 4-butylamino-2,2,6,6-tetramethylpiperidine (CAS Reg. No. [136504-96-6]); N-(2,2,6,6-tetramethyl-4-piperidyl)-n-dodecylsuccinimid, N-(1,2,2,6,6-pentamethyl-4-piperidyl)-n-dodecylsuccinimid, 2-undecyl-7,7,9,9-tetramethyl-1-oxa-3,8-diaza-4-oxo-spiro[4,5]decane, a reaction product of 7,7,9,9-tetramethyl-2-cycloundecyl-1-oxa-3,8-diaza-4-oxospiro[4,5]decane und epichlorohydrin, 1,1-bis(1,2,2,6,6-pentamethyl-4-piperidylloxycarbonyl)-2-(4-methoxyphenyl)ethene, N,N'-bis-formyl-N,N'-bis(2,2,6,6-tetramethyl-4-piperidyl)hexamethylenediamine, diester of 4-methoxy-methylene-malonic acid with 1,2,2,6,6-pentamethyl-4-hydroxypiperidine, poly[methylpropyl-3-oxy-4-(2,2,6,6-tetramethyl-4-piperidyl)]siloxane, reaction product of maleic acid anhydride-a-olefin-copolymer with 2,2,6,6-tetramethyl-4-aminopiperidine or 1,2,2,6,6-pentamethyl-4-aminopiperidine.

The polyolefin composition may contain further additives. Examples are given below.

1 Antioxidants

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1.1. Alkylated monophenols, for example 2,6-di-tert-butyl-4-methylphenol, 2-tert-butyl-4,6-di-methylphenol, 2,6-di-tert-butyl-4-ethylphenol, 2,6-di-tert-butyl-4-n-butylphenol, 2,6-di-tert-butyl-4-isobutylphenol, 2,6-dicyclopentyl-4-methylphenol, 2-(α -methylcyclohexyl)-4,6-dimethylphenol, 2,6-dioctadecyl-4-methylphenol, 2,4,6-tricyclohexylphenol, 2,6-di-tert-butyl-4-methoxymethylphenol, nonylphenols which are linear or branched in the side chains, for example, 2,6-di-nonyl-4-methylphenol, 2,4-dimethyl-6-(1'-methylundec-1'-yl)phenol, 2,4-dimethyl-6-(1'-methylheptadec-1'-yl)phenol, 2,4-dimethyl-6-(1'-methyltridec-1'-yl)phenol and mixtures thereof.

1.2. Alkylthiomethylphenols, for example 2,4-dioctylthiomethyl-6-tert-butylphenol, 2,4-dioctylthiomethyl-6-methylphenol, 2,4-dioctylthiomethyl-6-ethylphenol, 2,6-di-dodecylthiomethyl-4-nonylphenol.

1.3. Hydroquinones and alkylated hydroquinones, for example 2,6-di-tert-butyl-4-methoxyphenol, 2,5-di-tert-butylhydroquinone, 2,5-di-tert-amylhydroquinone, 2,6-diphenyl-4-octadecyloxyphenol, 2,6-di-tert-butylhydroquinone, 2,5-di-tert-butyl-4-hydroxyanisole, 3,5-di-tert-butyl-4-hydroxyanisole, 3,5-di-tert-butyl-4-hydroxyphenyl stearate, bis-(3,5-di-tert-butyl-4-hydroxyphenyl) adipate.

1.4. Tocopherols, for example α -tocopherol, β -tocopherol, γ -tocopherol, δ -tocopherol and mixtures thereof (Vitamin E).

1.5. Hydroxylated thiodiphenyl ethers, for example 2,2'-thiobis(6-tert-butyl-4-methylphenol), 2,2'-thiobis(4-octylphenol), 4,4'-thiobis(6-tert-butyl-3-methylphenol), 4,4'-thiobis(6-tert-butyl-2-methylphenol), 4,4'-thiobis-(3,6-di-sec-amylphenol), 4,4'-bis(2,6-dimethyl-4-hydroxyphenyl)disulfide.

1.6. Alkylidenebisphenols, for example 2,2'-methylenebis(6-tert-butyl-4-methylphenol), 2,2'-methylenebis(6-tert-butyl-4-ethylphenol), 2,2'-methylenebis[4-methyl-6-(α -methylcyclohexyl)-phenol], 2,2'-methylenebis(4-methyl-6-cyclohexylphenol), 2,2'-methylenebis(6-nonyl-4-methylphenol), 2,2'-methylenebis(4,6-di-tert-butylphenol), 2,2'-ethylidenebis(4,6-di-tert-butylphenol), 2,2'-ethylidenebis(6-tert-butyl-4-isobutylphenol), 2,2'-methylenebis[6-(α -methylbenzyl)-4-nonylphenol], 2,2'-methylenebis[6-(α , α -dimethylbenzyl)-4-nonylphenol], 4,4'-methylenebis(2,6-di-tert-butylphenol), 4,4'-methylenebis(6-tert-butyl-2-methylphenol), 1,1-bis(5-

tert-butyl-4-hydroxy-2-methylphenyl)butane, 2,6-bis(3-tert-butyl-5-methyl-2-hydroxybenzyl)-4-methylphenol, 1,1,3-tris(5-tert-butyl-4-hydroxy-2-methylphenyl)butane, 1,1-bis(5-tert-butyl-4-hydroxy-2-methyl-phenyl)-3-n-dodecylmercaptopbutane, ethylene glycol bis[3,3-bis(3'-tert-butyl-4'-hydroxyphenyl)butyrate], bis(3-tert-butyl-4-hydroxy-5-methyl-phenyl)dicyclopentadiene, bis[2-(3'-tert-butyl-2'-hydroxy-5'-methylbenzyl)-6-tert-butyl-4-methylphenyl]terephthalate, 1,1-bis-(3,5-dimethyl-2-hydroxyphenyl)butane, 2,2-bis-(3,5-di-tert-butyl-4-hydroxyphenyl)propane, 2,2-bis-(5-tert-butyl-4-hydroxy-2-methylphenyl)-4-n-dodecylmercaptopbutane, 1,1,5,5-tetra-(5-tert-butyl-4-hydroxy-2-methylphenyl)pentane.

1.7. O-, N- and S-benzyl compounds, for example 3,5,3',5'-tetra-tert-butyl-4,4'-dihydroxydi-benzyl ether, octadecyl-4-hydroxy-3,5-dimethylbenzylmercaptoacetate, tridecyl-4-hydroxy-3,5-di-tert-butylbenzylmercaptoacetate, tris(3,5-di-tert-butyl-4-hydroxybenzyl)amine, bis(4-tert-butyl-3-hydroxy-2,6-dimethylbenzyl)dithiophthalate, bis(3,5-di-tert-butyl-4-hydroxybenzyl)sulfide, isoctyl-3,5-di-tert-butyl-4-hydroxybenzylmercaptoacetate.

1.8. Hydroxybenzylated malonates, for example dioctadecyl-2,2-bis-(3,5-di-tert-butyl-2-hydroxybenzyl)-malonate, di-octadecyl-2-(3-tert-butyl-4-hydroxy-5-methylbenzyl)-malonate, di-dodecylmercaptoethyl-2,2-bis-(3,5-di-tert-butyl-4-hydroxybenzyl)malonate, bis[4-(1,1,3,3-tramethylbutyl)phenyl]-2,2-bis(3,5-di-tert-butyl-4-hydroxybenzyl)malonate.

1.9. Aromatic hydroxybenzyl compounds, for example 1,3,5-tris-(3,5-di-tert-butyl-4-hydroxybenzyl)-2,4,6-trimethylbenzene, 1,4-bis(3,5-di-tert-butyl-4-hydroxybenzyl)-2,3,5,6-tetramethylbenzene, 2,4,6-tris(3,5-di-tert-butyl-4-hydroxybenzyl)phenol.

1.10. Triazine Compounds, for example 2,4-bis(octylmercapto)-6-(3,5-di-tert-butyl-4-hydroxyanilino)-1,3,5-triazine, 2-octylmercapto-4,6-bis(3,5-di-tert-butyl-4-hydroxyanilino)-1,3,5-triazine, 2-octylmercapto-4,6-bis(3,5-di-tert-butyl-4-hydroxyphenoxy)-1,3,5-triazine, 2,4,6-tris(3,5-di-tert-butyl-4-hydroxyphenoxy)-1,2,3-triazine, 1,3,5-tris-(3,5-di-tert-butyl-4-hydroxybenzyl)isocyanurate, 1,3,5-tris(4-tert-butyl-3-hydroxy-2,6-dimethylbenzyl)isocyanurate, 2,4,6-tris(3,5-di-tert-butyl-4-hydroxyphenylethyl)-1,3,5-triazine, 1,3,5-tris(3,5-di-tert-butyl-4-hydroxyphenylpropionyl)-hexahydro-1,3,5-triazine, 1,3,5-tris(3,5-dicyclohexyl-4-hydroxybenzyl)isocyanurate.

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1.11. Benzylphosphonates, for example dimethyl-2,5-di-tert-butyl-4-hydroxybenzylphosphonate, diethyl-3,5-di-tert-butyl-4-hydroxybenzylphosphonate, dioctadecyl3,5-di-tert-butyl-4-hydroxybenzylphosphonate, dioctadecyl-5-tert-butyl-4-hydroxy-3-methylbenzylphosphonate, the calcium salt of the monoethyl ester of 3,5-di-tert-butyl-4-hydroxybenzylphosphonic acid.

1.12. Acylaminophenols, for example 4-hydroxylauranilide, 4-hydroxystearanilide, octyl N-(3,5-di-tert-butyl-4-hydroxyphenyl)carbamate.

1.13. Esters of β -(3,5-di-tert-butyl-4-hydroxyphenyl)propionic acid with mono- or polyhydric alcohols, e.g. with methanol, ethanol, n-octanol, i-octanol, octadecanol, 1,6-hexanediol, 1,9-nonenediol, ethylene glycol, 1,2-propanediol, neopentyl glycol, thiodiethylene glycol, diethylene glycol, triethylene glycol, pentaerythritol, tris(hydroxyethyl) isocyanurate, N,N'-bis(hydroxyethyl)oxamide, 3-thiaundecanol, 3-thiapentadecanol, trimethylhexanediol, trimethylolpropane, 4-hydroxymethyl-1-phospha-2,6,7-trioxabicyclo[2.2.2]octane.

1.14. Esters of β -(5-tert-butyl-4-hydroxy-3-methylphenyl)propionic acid with mono- or polyhydric alcohols, e.g. with methanol, ethanol, n-octanol, i-octanol, octadecanol, 1,6-hexanediol, 1,9-nonenediol, ethylene glycol, 1,2-propanediol, neopentyl glycol, thiodiethylene glycol, diethylene glycol, triethylene glycol, pentaerythritol, tris(hydroxyethyl) isocyanurate, N,N'-bis(hydroxyethyl)oxamide, 3-thiaundecanol, 3-thiapentadecanol, trimethylhexanediol, trimethylolpropane, 4-hydroxymethyl-1-phospha-2,6,7-trioxabicyclo[2.2.2]octane.

1.15. Esters of β -(3,5-dicyclohexyl-4-hydroxyphenyl)propionic acid with mono- or polyhydric alcohols, e.g. with methanol, ethanol, octanol, octadecanol, 1,6-hexanediol, 1,9-nonenediol, ethylene glycol, 1,2-propanediol, neopentyl glycol, thiodiethylene glycol, diethylene glycol, triethylene glycol, pentaerythritol, tris(hydroxyethyl)isocyanurate, N,N'-bis(hydroxyethyl)oxamide, 3-thiaundecanol, 3-thiapentadecanol, trimethylhexanediol, trimethylolpropane, 4-hydroxymethyl-1-phospha-2,6,7-trioxabicyclo[2.2.2]octane.

1.16. Esters of 3,5-di-tert-butyl-4-hydroxyphenyl acetic acid with mono- or polyhydric alcohols, e.g. with methanol, ethanol, octanol, octadecanol, 1,6-hexanediol, 1,9-nonenediol, ethylene glycol, 1,2-propanediol, neopentyl glycol, thiodiethylene glycol, diethylene glycol, triethylene glycol, pentaerythritol, tris(hydroxyethyl)isocyanurate, N,N'-bis(hydroxyethyl)ox-

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amide, 3-thiaundecanol, 3-thiapentadecanol, trimethylhexanediol, trimethylopropane, 4-hydroxymethyl-1-phospha-2,6,7-trioxabicyclo[2.2.2]octane.

1.17. Amides of β -(3,5-di-tert-butyl-4-hydroxyphenyl)propionic acid e.g. N,N'-bis(3,5-di-tert-butyl-4-hydroxyphenylpropionyl)hexamethylenediamide, N,N'-bis(3,5-di-tert-butyl-4-hydroxyphenylpropionyl)trimethylenediamide, N,N'-bis(3,5-di-tert-butyl-4-hydroxyphenylpropionyl)-hydrazide, N,N'-bis[2-(3-[3,5-di-tert-butyl-4-hydroxyphenyl]propionyloxy)ethyl]oxamide (Naugard[®]XL-1 supplied by Uniroyal).

1.18. Ascorbic acid (vitamin C)

1.19. Aminic antioxidants, for example N,N'-di-isopropyl-p-phenylenediamine, N,N'-di-sec-butyl-p-phenylenediamine, N,N'-bis(1,4-dimethylpentyl)-p-phenylenediamine, N,N'-bis(1-ethyl-3-methylpentyl)-p-phenylenediamine, N,N'-bis(1-methylheptyl)-p-phenylenediamine, N,N'-dicyclohexyl-p-phenylenediamine, N,N'-diphenyl-p-phenylenediamine, N,N'-bis(2-naphthyl)-p-phenylenediamine, N-isopropyl-N'-phenyl-p-phenylenediamine, N-(1,3-dimethylbutyl)-N'-phenyl-p-phenylenediamine, N-(1-methylheptyl)-N'-phenyl-p-phenylenediamine, N-cyclohexyl-N'-phenyl-p-phenylenediamine, 4-(p-toluenesulfamoyl)diphenylamine, N,N'-dimethyl-N,N'-di-sec-butyl-p-phenylenediamine, diphenylamine, N-allyldiphenylamine, 4-isopropoxy-diphenylamine, N-phenyl-1-naphthylamine, N-(4-tert-octylphenyl)-1-naphthylamine, N-phenyl-2-naphthylamine, octylated diphenylamine, for example p,p'-di-tert-octyldiphenylamine, 4-n-butylaminophenol, 4-butyrylaminophenol, 4-nonanoylaminophenol, 4-dodecanoylemino-phenol, 4-octadecanoylaminophenol, bis(4-methoxyphenyl)amine, 2,6-di-tert-butyl-4-dimethylaminomethylphenol, 2,4'-diaminodiphenylmethane, 4,4'-diaminodiphenylmethane, N,N,N',N'-tetramethyl-4,4'-diaminodiphenylmethane, 1,2-bis[(2-methylphenyl)amino]ethane, 1,2-bis(phenylamino)propane, (o-tolyl)biguanide, bis[4-(1',3'-dimethylbutyl)phenyl]amine, tert-octylated N-phenyl-1-naphthylamine, a mixture of mono- and dialkylated tert-butyl/tert-octyldiphenylamines, a mixture of mono- and dialkylated nonyldiphenylamines, a mixture of mono- and dialkylated dodecyldiphenylamines, a mixture of mono- and dialkylated isopropyl/isohexyldiphenylamines, a mixture of mono- und dialkylated tert-butyldiphenylamines, 2,3-dihydro-3,3-dimethyl-4H-1,4-benzothiazine, phenothiazine, a mixture of mono- und dialkylated tert-butyl/tert-octylphenothiazines, a mixture of mono- und dialkylated tert-octyl-phenothiazines, N-allylphenothiazin, N,N,N',N'-tetraphenyl-1,4-diaminobut-2-ene, N,N-bis-

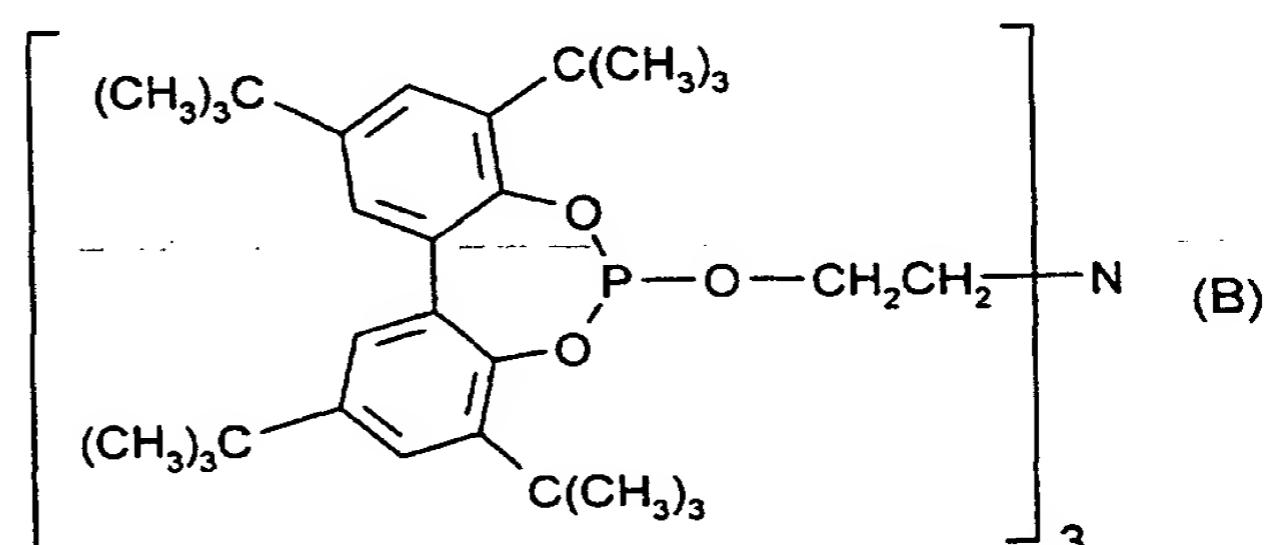
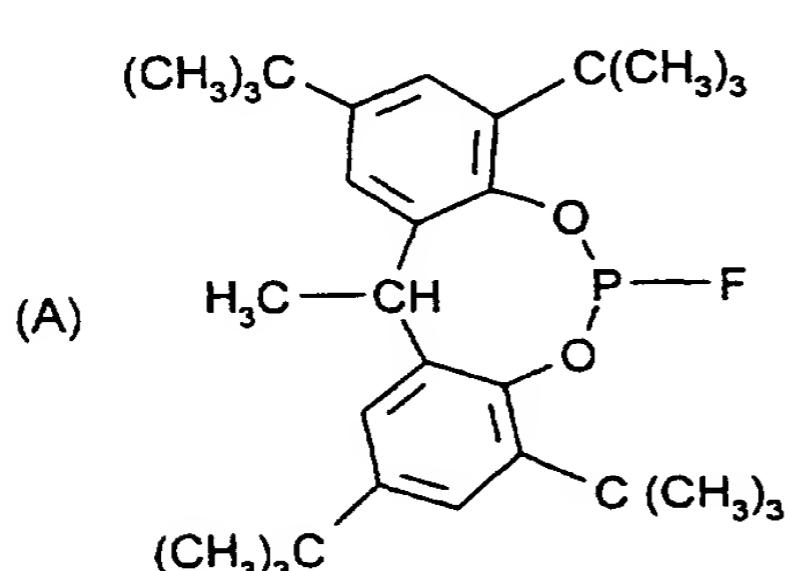
- 21 -

(2,2,6,6-tetramethyl-piperid-4-yl-hexamethylenediamine, bis(2,2,6,6-tetramethylpiperid-4-yl)-sebacate, 2,2,6,6-tetramethylpiperidin-4-one, 2,2,6,6-tetramethylpiperidin-4-ol.

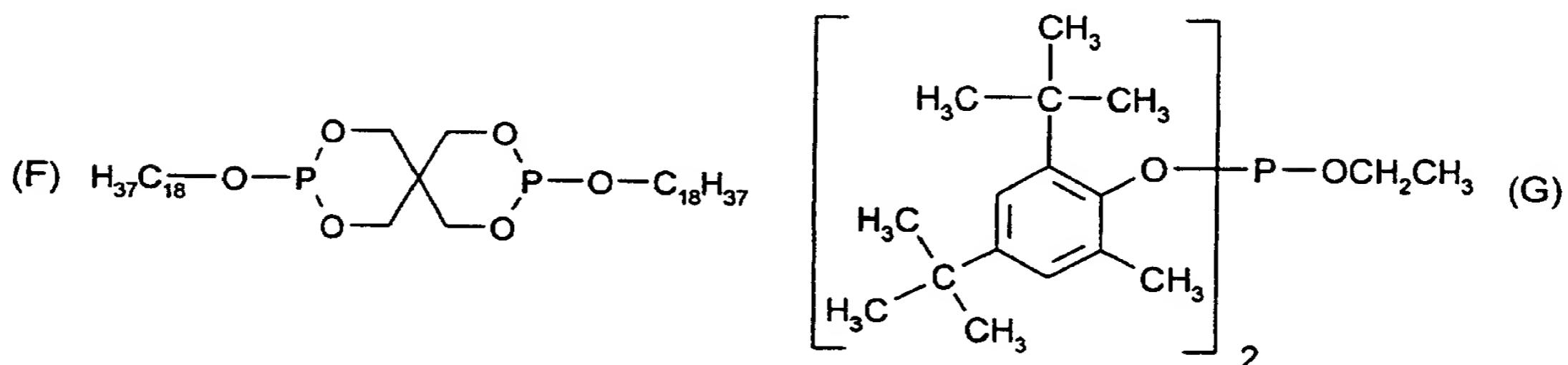
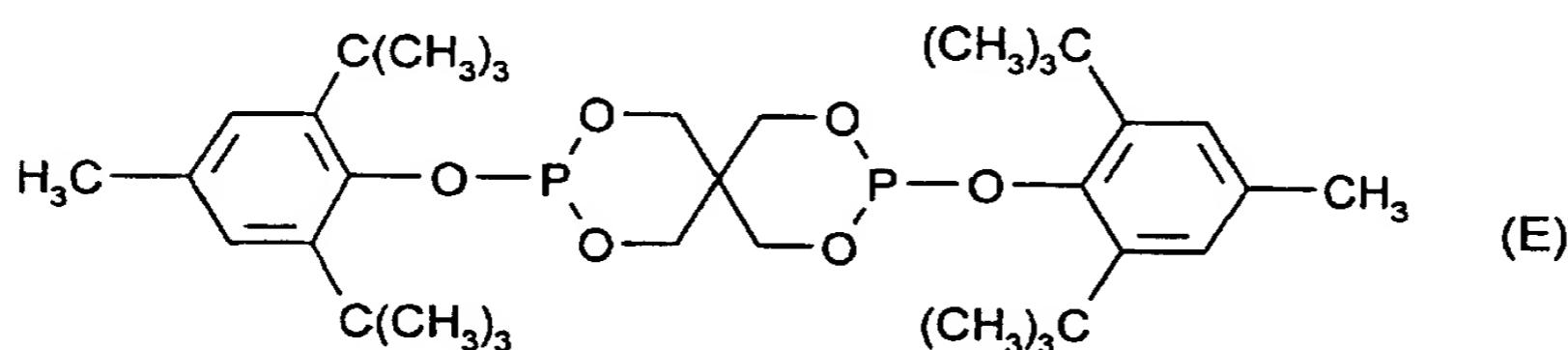
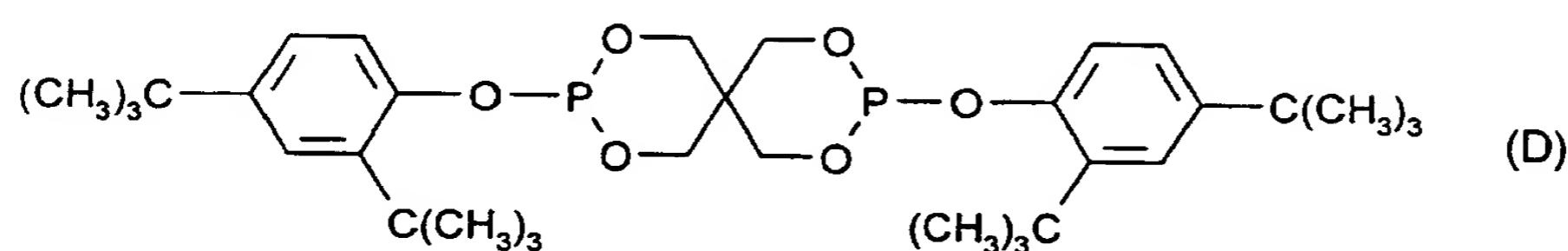
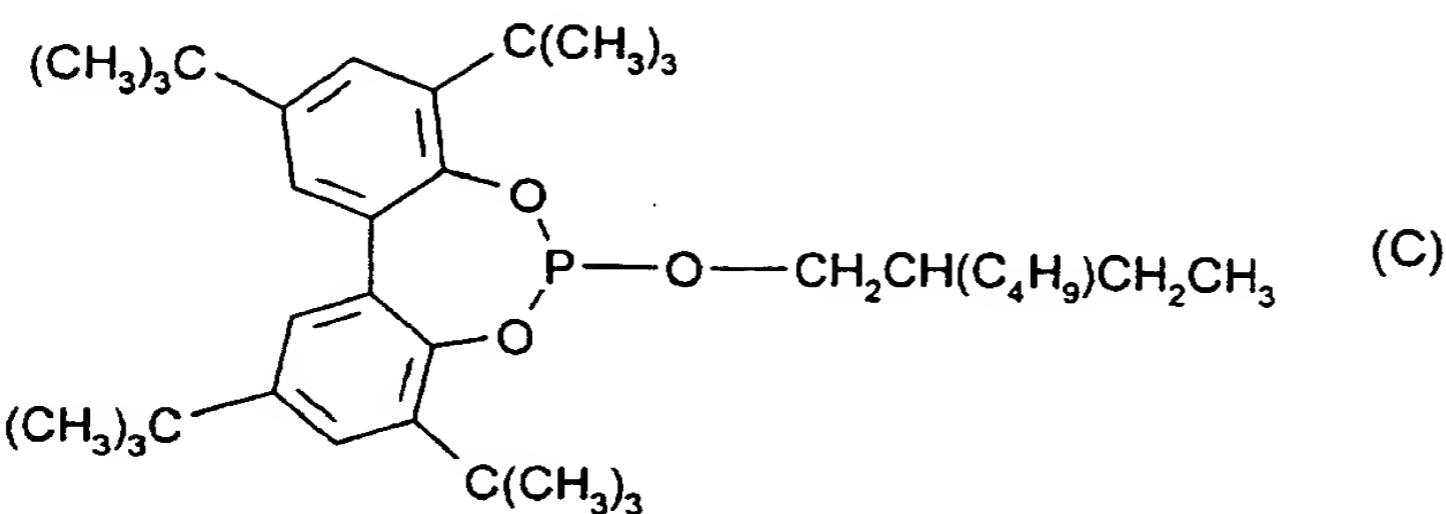
2. Phosphites and phosphonites, for example triphenyl phosphite, diphenyl alkyl phosphites, phenyl dialkyl phosphites, tris(nonylphenyl) phosphite, trilauryl phosphite, trioctadecyl phosphite, distearyl pentaerythritol diphosphite, tris(2,4-di-tert-butylphenyl) phosphite, diisodecyl pentaerythritol diphosphite, bis(2,4-di-tert-butylphenyl) pentaerythritol diphosphite, bis(2,6-di-tert-butyl-4-methylphenyl)-pentaerythritol diphosphite, diisodecyloxy pentaerythritol diphosphite, bis(2,4-di-tert-butyl-6-methylphenyl)pentaerythritol diphosphite, bis(2,4,6-tris(tert-butylphenyl)pentaerythritol diphosphite, tristearyl sorbitol triphosphite, tetrakis(2,4-di-tert-butylphenyl) 4,4'-biphenylene diphosphonite, 6-isooctyloxy-2,4,8,10-tetra-tert-butyl-12H-dibenz[d,g]-1,3,2-dioxaphosphocin, 6-fluoro-2,4,8,10-tetra-tert-butyl-12-methyl-dibenz[d,g]-1,3,2-dioxaphosphocin, bis(2,4-di-tert-butyl-6-methylphenyl) methyl phosphite, bis(2,4-di-tert-butyl-6-methylphenyl) ethyl phosphite, 2,2',2"-nitrilo[triethyltris(3,3',5,5'-tetra-tert-butyl-1,1'-biphenyl-2,2'-diyl)phosphite], 2-ethylhexyl(3,3',5,5'-tetra-tert-butyl-1,1'-biphenyl-2,2'-diyl)phosphite.

Especially preferred are the following phosphites:

Tris(2,4-di-tert-butylphenyl) phosphite (Irgafos[®]168, Ciba-Geigy), tris(nonylphenyl) phosphite,



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3. Hydroxylamines, for example, N,N-dibenzylhydroxylamine, N,N-diethylhydroxylamine, N,N-dioctylhydroxylamine, N,N-dilaurylhydroxylamine, N,N-ditetradecylhydroxylamine, N,N-dihexadecylhydroxylamine, N,N-dioctadecylhydroxylamine, N-hexadecyl-N-octadecylhydroxylamine, N-heptadecyl-N-octadecylhydroxylamine, N,N-dialkylhydroxylamine derived from hydrogenated tallow amine.

4. Nitrones, for example, N-benzyl-alpha-phenyl-nitron, N-ethyl-alpha-methyl-nitron, N-octyl-alpha-heptyl-nitron, N-lauryl-alpha-undecyl-nitron, N-tetradecyl-alpha-tridecyl-nitron, N-hexadecyl-alpha-pentadecyl-nitron, N-octadecyl-alpha-heptadecyl-nitron, N-hexadecyl-

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alpha-heptadecyl-nitrone, N-octadecyl-alpha-pentadecyl-nitrone, N-heptadecyl-alpha-heptadecyl-nitrone, N-octadecyl-alpha-hexadecyl-nitrone, nitrone derived from N,N-dialkylhydroxylamine derived from hydrogenated tallow amine.

5. Nucleating agents, for example, inorganic substances such as talcum, metal oxides such as titanium dioxide or magnesium oxide, phosphates, carbonates or sulfates of, preferably, alkaline earth metals; organic compounds such as mono- or polycarboxylic acids and the salts thereof, e.g. 4-tert-butylbenzoic acid, adipic acid, diphenylacetic acid, sodium succinate or sodium benzoate; polymeric compounds such as ionic copolymers (ionomers).

6. Fillers and reinforcing agents, for example, calcium carbonate, silicates, glass fibres, glass bulbs, asbestos, talc, kaolin, mica, barium sulfate, metal oxides and hydroxides, carbon black, graphite, wood flour and flours or fibers of other natural products, synthetic fibers.

7. Other additives, for example, plasticisers, lubricants, emulsifiers, pigments, rheology additives, catalysts, flow-control agents, optical brighteners, flameproofing agents, antistatic agents and blowing agents.

8. Benzofuranones and indolinones, for example those disclosed in U.S. 4,325,863; U.S. 4,338,244; U.S. 5,175,312; U.S. 5,216,052; U.S. 5,252,643; DE-A-4316611; DE-A-4316622; DE-A-4316876; EP-A-0589839 or EP-A-0591102 or 3-[4-(2-acetoxyethoxy)phenyl]-5,7-di-tert-butyl-benzofuran-2-one, 5,7-di-tert-butyl-3-[4-(2-stearoyloxyethoxy)phenyl]benzofuran-2-one, 3,3'-bis[5,7-di-tert-butyl-3-(4-[2-hydroxyethoxy]phenyl)benzofuran-2-one], 5,7-di-tert-butyl-3-(4-ethoxyphenyl)benzofuran-2-one, 3-(4-acetoxy-3,5-dimethylphenyl)-5,7-di-tert-butyl-benzofuran-2-one, 3-(3,5-dimethyl-4-pivaloyloxyphenyl)-5,7-di-tert-butyl-benzofuran-2-one, 3-(3,4-dimethylphenyl)-5,7-di-tert-butyl-benzofuran-2-one, 3-(2,3-dimethylphenyl)-5,7-di-tert-butyl-benzofuran-2-one.

A further subject of the invention is the use of a mixture of UV-absorbers as defined above for the stabilization of polyolefins.

The following examples illustrate the invention.

Example 1: Light stabilization of PP homopolymer films

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100 parts of unstabilized polypropylene powder (melt flow index 3.2 g/10 minutes, 230°C / 2160 g) are homogenized at 200°C for 10 minutes in a Brabender plastograph with 0.05 parts of pentaerythrityl-tetrakis-3-(3,5-di-tert.butyl-4-hydroxyphenyl)-propionate, 0.05 parts of tris-(2,4-di-tert.butylphenyl)-phosphite, 0.1 parts of Ca stearate and the amount of light stabilizer indicated in table 1. The material thus obtained is compression molded in a laboratory press between two aluminum foils for 6 minutes at 260°C to a 0.5 mm thick film which is cooled immediately to room temperature in a water-cooled press. Samples of 60 x 25 mm are cut out of these 0.5 mm films and exposed in a WEATHER-OMETER Ci 65 (black panel temperature 63±2°C, without water-spraying)

Periodically, these samples are removed from the exposure apparatus and their carbonyl content is measured with an infrared spectrophotometer.

The exposure time corresponding to formation of a carbonyl absorbance of 0.1 is a measure for the stabilizing efficiency of the light stabilizer. The values obtained are summarized in the following table (Table 1).

Table 1: Performance in PP Homopolymer 0.5 mm Thick Films

Stabilization	T0.1: (h) WEATHER- OMETER Ci 65	Calculated additive effect of combinations
Control	120	-
0.2% CHIMASSORB 81	410	-
0.2% TINUVIN 326	430	-
0.2% TINUVIN 1577	570	-
0.2% TINUVIN 315	905	-
0.1% CHIMASSORB 81 + 0.1% TINUVIN 1577	650	490
0.1% CHIMASSORB 81 + 0.1% TINUVIN 315	770	657
0.1% TINUVIN 326 + 0.1% TINUVIN 315	705	667
0.1% TINUVIN 1577 + 0.1% TINUVIN 315	810	737

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Example 2: Light stabilization of PP homopolymer films

100-parts of unstabilized polypropylene powder (melt flow index 12 g/10 minutes, 230°C / 2160 g) are homogenized at 200°C for 10 minutes in a Brabender plastograph with 0.05 parts of pentaerythrityl-tetrakis-3-(3,5-ditert.butyl-4-hydroxyphenyl)-propionate, 0.05 parts of tris-(2,4-di-tert.butylphenyl)-phosphite, 0.1 parts of Ca stearate and the amount of light stabilizer indicated in table 2. The material thus obtained is compression molded in a laboratory press between two aluminum foils for 6 minutes at 260°C to a 0.1 mm thick film, which is cooled immediately to room temperature in a water-cooled press. Samples of 60 x 25 mm are cut out of these 0.1 mm films and exposed in a XENOTEST 1200 (black panel temperature 53±2°C, without water-spraying).

Periodically, these samples are removed from the exposure apparatus and their carbonyl content is measured with an infrared spectrophotometer.

The exposure time corresponding to formation of a carbonyl absorbance of 0.1 is a measure for the stabilizing efficiency of the light stabilizer. The values obtained are summarized in the following table (Table 2).

Table 2: Light Stabilization of PP Homopolymer Films

Stabilization	T0.1: (h) XENOTEST 1200	Calculated additive effect of combinations
Control	300	-
0.6% CHIMASSORB 81	3120	-
0.6% TINUVIN 327	440	-
0.6% TINUVIN 315	1090	-
0.3% CHIMASSORB 81 +0.3% TINUVIN 315	2650	2105
0.3% TINUVIN 327+0.3% TINUVIN 315	940	765
0.2% CHIMASSORB 81 +0.2% TINUVIN 327 +0.2% TINUVIN 315	1870	1550
0.4% CHIMASSORB 81 +0.1% TINUVIN 327 +0.1% TINUVIN 315	2925	2335

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0.3% CHIMASSORB 81 +0.15% TINUVIN 327	2720	2030
+0.15% TINUVIN 315		

Example 3: Light stabilization of PE-HD films (Phillips-type)

100 parts of a Phillips-type high density polyethylene powder (density: 0.965, melt flow index 6.0 g/10 minutes, 190°C / 2160 g) stabilized with 0.05 parts of pentaerythrityl-tetrakis-3-(3,5-ditert.butyl-4-hydroxyphenyl)-propionate and 0.1 parts of Ca stearate are homogenized at 180°C for 10 minutes in a Brabender plastograph with the amount of light stabilizer and coadditives indicated in table 3. Similarly 100 parts of a Ziegler-type high density polyethylene powder (density: 0.961, melt flow index 6.0 g/10 minutes, 190°C/2160 g) stabilized with 0.05 parts of pentaerythrityl-tetrakis-3-(3,5-ditert.butyl-4-hydroxyphenyl)-propionate, 0.05 parts tris-(2,4-di-tert.butylphenyl)-phosphite and 0.1 parts of Ca stearate are homogenized at 180°C for 10 minutes in a Brabender plastograph with the amount of light stabilizer and coadditives indicated in table 3a.

The material thus obtained is compression molded in a laboratory press between two aluminum foils for 6 minutes at 210°C to a 0.2 mm thick film which is cooled immediately to room temperature in a water-cooled press. Samples of 60 x 25 mm are cut out of these 0.2 mm films and exposed in a XENOTEST 1200 (black panel temperature 53±2°C, without water spraying).

Periodically, these samples are removed from the exposure apparatus and their carbonyl content is measured with an infrared spectrophotometer.

The exposure time corresponding to formation of a carbonyl absorbance of 0.1 is a measure for the stabilizing efficiency of the light stabilizer. The values obtained are summarized in the following tables 3 and 3a.

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Table 3: Light Stabilization of 0.2 mm Thick PE-HD Films (Phillips Type)

Stabilization	T _{0.1} : (h) XENOTEST	Calculated additive effect of combinations
	1200	
Control	360	-
0.3% CHIMASSORB 81	2230	-
0.3% TINUVIN 327	1240	-
0.3% TINUVIN 315	2020	-
0.15% CHIMASSORB 81 + 0.15% TINUVIN 327	3000	1735
0.15% CHIMASSORB 81 + 0.15% TINUVIN 315	2650	2125
0.15% TINUVIN 327 + 0.15% TINUVIN 315	3060	1630
0.1% CHIMASSORB 81 + 0.1% TINUVIN 327 + 0.1% TINUVIN 315	3150	1830

Table 3a:

Light stabilization of 0.2 mm Thick PE-HD Films (Ziegler Type)

Stabilization	T _{0.1} : (h) XENOTEST	Calculated additive effect of combinations
	1200	
Comparative Experiment		
Control	190	-
0.3% CHIMASSORB 81	1540	-
0.3% TINUVIN 327	700	-
0.15% CHIMASSORB 81 + 0.15% TINUVIN 327	940	1120

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Example 4: Light stabilization of PE-LD films

100 parts of unstabilized low density polyethylene powder (density: 0.918, melt flow index 0.3 g/10 minutes at 190°C / 2160 g) are homogenized at 180°C for 10 minutes in a Brabender plastograph with 0.02 parts of octadecyl 3-(3,5-di-tert.-butyl-4-hydroxyphenyl)-propionate, 0.08 parts of tris-(2,4-di-tert.butylphenyl)-phosphite and the amount of light stabilizer indicated in the table. The material thus obtained is compression molded in a laboratory press between two aluminum foils for 6 minutes at 170°C to a 0.5 mm thick film which is quenched immediately in cold water. Samples of 60 x 25 mm are cut out of these 0.5 mm films and exposed in a WEATHER-OMETER Ci 65 (black panel temperature 63±2°C, without water-spraying).

Periodically, these samples are removed from the exposure apparatus and their carbonyl content is measured with an infrared spectrophotometer.

The exposure time corresponding to formation of a carbonyl absorbance of 0.1 is a measure for the stabilizing efficiency of the light stabilizer. The values obtained are summarized in the following table.

Table 4: Light Stabilization of PE-LD Films

Stabilization	T0.1: (h)	Calculated
	WEATHER-	additive
	OMETER Ci 65	effect of combinations
Control	150	-
0.1% CHIMASSORB 81	430	-
0.2% CHIMASSORB 81	665	-
0.1% TINUVIN 326	760	-
0.2% TINUVIN 326	1150	-
0.1% TINUVIN 1577	815	-
0.2% TINUVIN 1577	1100	-
0.1% TINUVIN 315	705	-
0.2% TINUVIN 315	2000	-
0.05% CHIMASSORB 81 +0.05% TINUVIN 1577	665	622
0.05% CHIMASSORB 81 +0.05% TINUVIN 315	780	567
0.05% TINUVIN 326 +0.05% TINUVIN 315	795	732
0.05% TINUVIN 1577 +0.05% TINUVIN 315	830	760

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0.1% CHIMASSORB 81 +0.1% TINUVIN 1577	920	882
0.1% CHIMASSORB 81 +0.1% TINUVIN 315	1660	1332

Example 5: Light stabilization of PE-LLD films

100 parts of unstabilized linear low density polyethylene (PE-LLD) powder (density: 0.920, melt flow index 1.0 g/10 minutes at 190°C/2160 g) are homogenized at 180°C for 10 minutes in a Brabender plastograph with 0.02 parts octadecyl 3-(3,5-di-tert.-butyl-4-hydroxyphenyl)-propionate, 0.08 parts tris-(2,4-di-tert.butylphenyl)-phosphite, 0.1 parts Ca stearate and the amounts light stabilizers indicated in table 5. Similarly 100 parts of unstabilized metallocene polyethylene powder (density: 0.905, 10% butene comonomer, melt flow index 1.4 g/10 minutes at 190°C/2160 g) are homogenized at 180°C for 10 minutes in a Brabender plastograph with 0.02 parts octadecyl 3-(3,5-di-tert.-butyl-4-hydroxyphenyl)-propionate, 0.08 parts tris-(2,4-di-tert.butylphenyl)-phosphite, 0.1 parts Ca stearate and the amounts light stabilizers indicated in table 5a (see also example 6). The material thus obtained is compression molded in a laboratory press between two aluminum foils for 6 minutes at 170°C to a 0.5-mm thick film, which is quenched immediately in cold water. Samples of 60 x 25 mm are cut out of these 0.5 mm films and exposed in a WEATHER-OMETER Ci 65 (black panel temperature 63±2°C, without water-spraying).

Periodically, these samples are removed from the exposure apparatus and their carbonyl content is measured with an infrared spectrophotometer.

The exposure time corresponding to formation of a carbonyl absorbance of 0.1 is a measure for the stabilizing efficiency of the light stabilizer. The values obtained are summarized in the following tables 5 and 5a.

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Table 5: Light Stabilization of PE-LLD Conventional Type

Stabilization	T0.1: (h) WEATHER- OMETER Ci 65	Calculated additive effect of combinations
Control	260	-
0.2% CHIMASSORB 81	2260	-
0.2% TINUVIN 326	1350	-
0.2% TINUVIN 1577	1600	-
0.2% TINUVIN 315	2840	-
0.1% CHIMASSORB 81 +0.1% TINUVIN 315	3300	2550
0.1% TINUVIN 326 + 0.1% TINUVIN 315	2820	2095
0.1% TINUVIN 1577 + 0.1% TINUVIN 315	2920	2220

Table 5a: Comparison Between PE-LLD Metallocene Type and Conventional PE-LLD for Combinations Benzophenon/Benzotriazol UV-Absorber (data partly from table 5 and 6)

Stabilization metallocene PE-LLD	T0.1: (h) WEATHER- OMETER Ci 65	Calculated additive effect of combinations
Control	365	-
0.2% CHIMASSORB 81	1920	-
0.2% TINUVIN 326	1480	-
0.1% CHIMASSORB 81 +0.1% TINUVIN 326	1880	1700

Comparative example with conventional PE-LLD

Control	260	-
0.2% CHIMASSORB 81	2260	-
0.2% TINUVIN 326	1350	-
0.1% CHIMASSORB 81 +0.1% TINUVIN 326	1390	1805

Example 6: Light stabilization of metallocene-PE films

100 parts of unstabilized metallocene polyethylene powder (density: 0.905, 10 % butene comonomer) are homogenized at 180°C for 10 minutes in a Brabender plastograph with 0.02 parts of octadecyl 3-(3,5-di-tert.-butyl-4-hydroxyphenyl)-propionate, 0.08 parts of tris-(2,4-di-tert.butylphenyl)-phosphite, 0.1 part of Ca stearate and the amount of light stabilizer indicated in the table. The material thus obtained is compression molded in a laboratory press between two aluminum foils for 6 minutes at 170°C to a 0.2 mm thick film which is quenched immediately in cold water. Samples of 60 x 25 mm are cut out of these 0.2 mm films and exposed in a WEATHER-OMETER Ci 65 (black panel temperature 63±2°C, without water-spraying).

Periodically, these samples are removed from the exposure apparatus and their carbonyl content is measured with an infrared spectrophotometer.

The exposure time corresponding to formation of a carbonyl absorbance of 0.1 is a measure for the stabilizing efficiency of the light stabilizer. The values obtained are summarized in the following table.

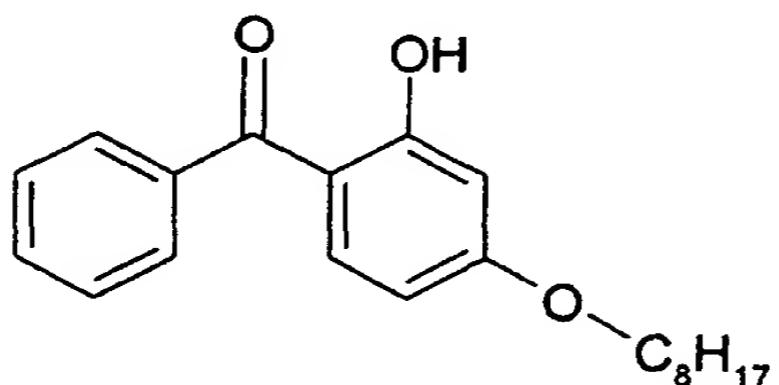
Table 6: Light Stabilization of Metallocene Polyethylene (m-PE-LD)

Stabilization	T0.1: (h) WEATHER- OMETER Ci 65	Calculated additive effect of combinations
Control	365	-
0.2% CHIMASSORB 81	1920	-
0.2% TINUVIN 326	1480	-
0.2% TINUVIN 1577	1800	-
0.2% TINUVIN 315	3380	-
0.1% CHIMASSORB 81 + 0.1% TINUVIN 326	1880	1700
0.1% CHIMASSORB 81 + 0.1% TINUVIN 1577	2060	1860
0.1% CHIMASSORB 81 + 0.1% TINUVIN 315	3440	2650

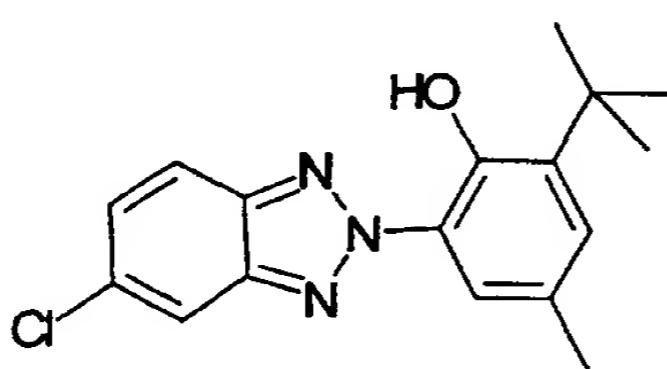
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All compounds used are commercial UV-absorbers from Ciba Specialty Chemicals Inc. and the trade names are registered trade marks.

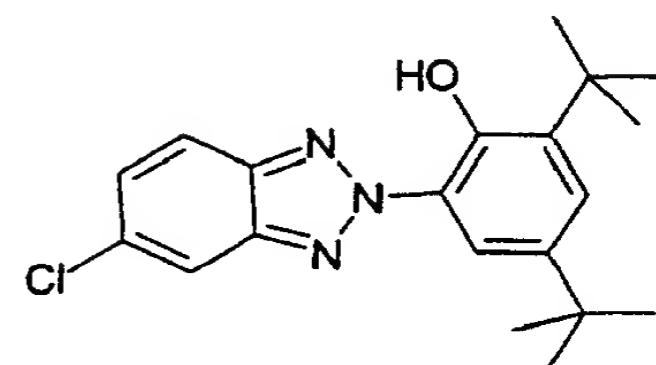
Chimassorb® 81



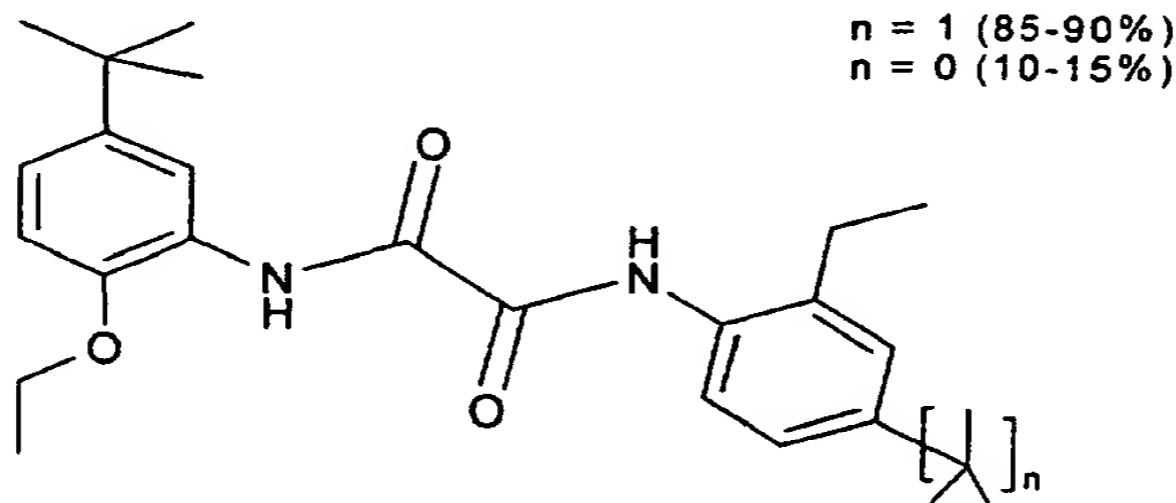
Tinuvin® 326



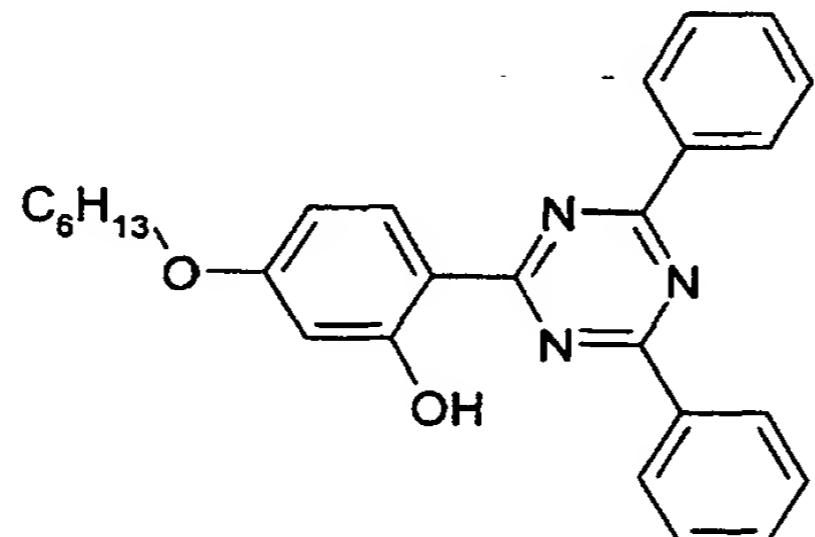
, Tinuvin® 327



Tinuvin® 315

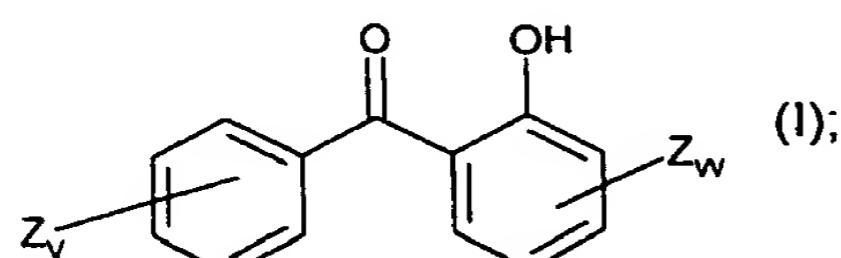


Tinuvin® 1577



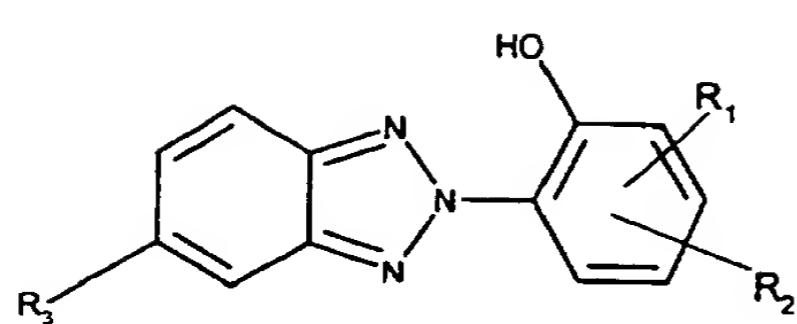
Claims

1. A polyolefin composition which comprises as UV absorber a mixture of
 - a) at least one hydroxybenzophenone and at least one 2-hydroxyphenylbenzotriazole with the proviso that the polyolefin is a high density polyethylene or a polypropylene of the "Phillips" type or a polyethylene or polypropylene of the metallocene type and no polyvinylpyridin is present;
 - b) at least one hydroxybenzophenone and at least one 2-hydroxyphenyltriazine, with the proviso that if the polyolefin is polypropylene, no polyvinylpyridin is present
 - c) at least one hydroxybenzophenone and at least one oxanilide;
 - d) at least one 2-hydroxyphenylbenzotriazole and at least one oxanilide;
 - e) at least one 2-hydroxyphenyltriazine and at least one oxanilide;
 - f) at least one hydroxybenzophenone, at least one 2-hydroxyphenylbenzotriazole and at least one oxanilide;
 - g) at least one hydroxybenzophenone, at least one 2-hydroxyphenylbenzotriazole and at least one 2-hydroxyphenyltriazine, with the proviso that if the polyolefin is polypropylene, no polyvinylpyridin is present;
 - h) at least one hydroxybenzophenone, at least one oxanilide and at least one 2-hydroxyphenyltriazine; or
 - i) at least one 2-hydroxyphenylbenzotriazole, at least one oxanilide and at least one 2-hydroxyphenyltriazine.
2. A polyolefin composition according to claim 1 wherein the polyolefin is polyethylene or polypropylene.
3. A polyolefin composition according to claim 1 wherein the hydroxybenzophenone is of formula I

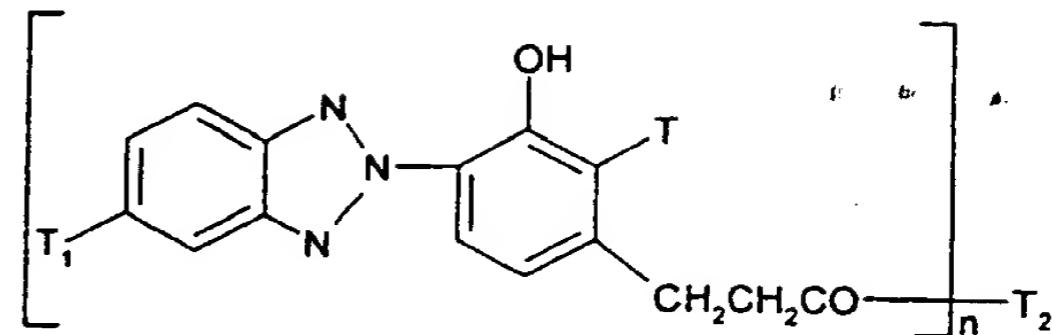


the 2-hydroxyphenylbenzotriazole is of formula IIa, IIb or IIc

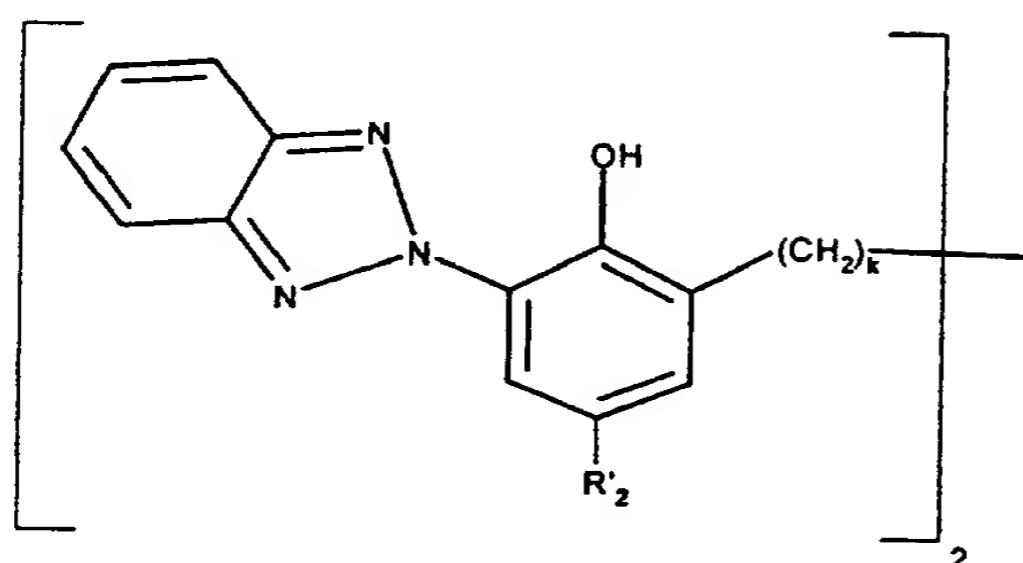
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(IIa),

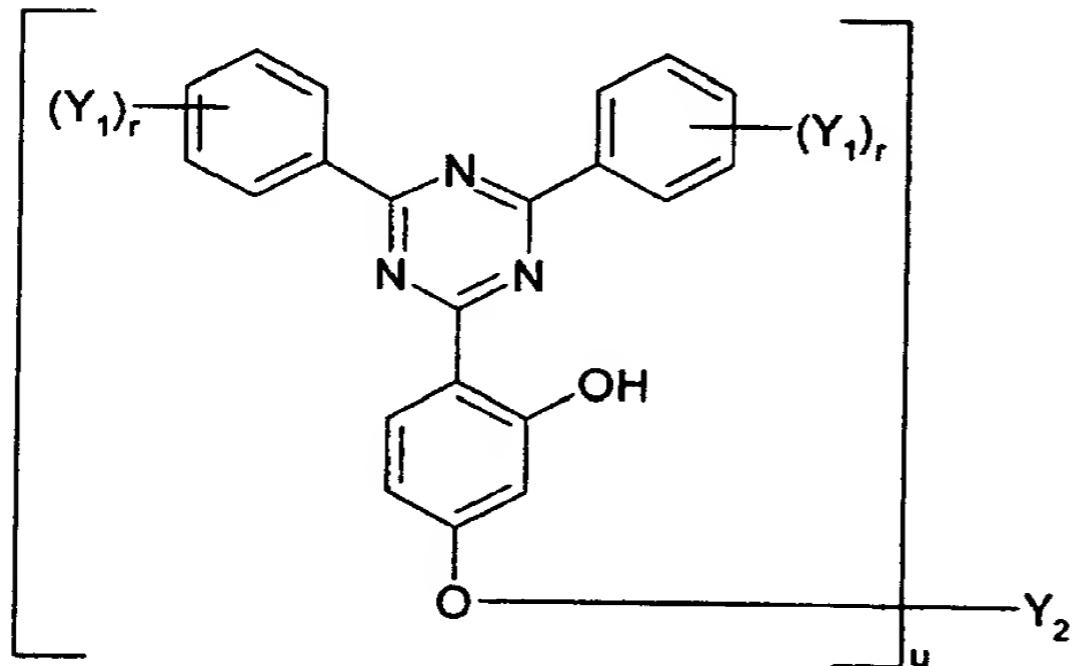


(IIb)



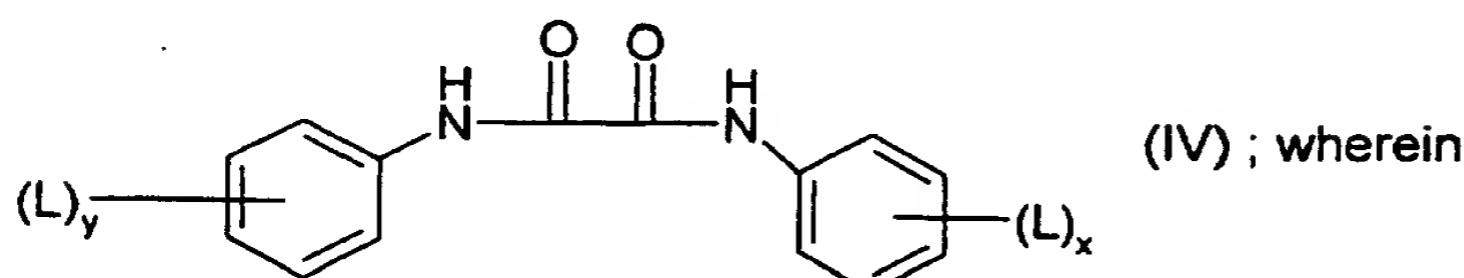
(IIc);

the 2-hydroxyphenyltriazine is of formula III



(III);

and the oxanilide is of formula (IV)



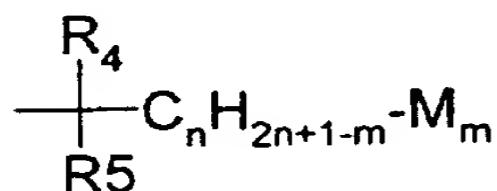
(IV); wherein

in the compounds of the formula (I) v is an integer from 1 to 3 and w is 1 or 2 and the

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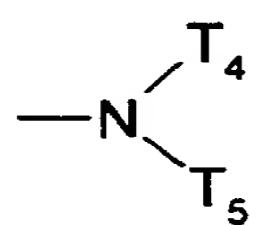
substituents Z independently of one another are hydrogen, halogen, hydroxyl or alkoxy having 1 to 12 carbon atoms;

in the compounds of the formula (IIa), R₁ is hydrogen, alkyl having 1 to 24 carbon atoms, phenylalkyl having 1 to 4 carbon atoms in the alkyl moiety, cycloalkyl having 5 to 8 carbon atoms or a radical of the formula

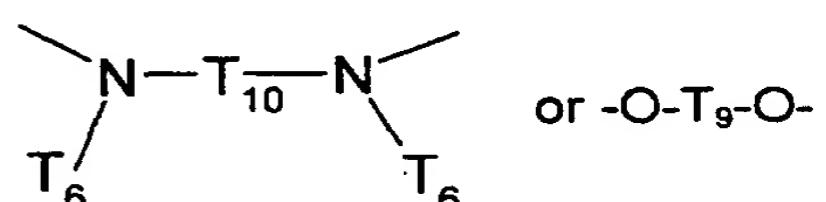


in which R₄ and R₅ independently of one another are alkyl having in each case 1 to 5 carbon atoms, or R₄, together with the radical C_nH_{2n+1-m}, forms a cycloalkyl radical having 5 to 12 carbon atoms, m is 1 or 2, n is an integer from 2 to 20 and M is a radical of the formula -COOR₆ in which R₆ is hydrogen, alkyl having 1 to 12 carbon atoms, alkoxyalkyl having in each case 1 to 20 carbon atoms in the alkyl moiety and in the alkoxy moiety or phenylalkyl having 1 to 4 carbon atoms in the alkyl moiety, R₂ is hydrogen, halogen, alkyl having 1 to 18 carbon atoms, and phenylalkyl having 1 to 4 carbon atoms in the alkyl moiety, and R₃ is hydrogen, chlorine, alkyl or alkoxy having in each case 1 to 4 carbon atoms or -COOR₆ in which R₆ is as defined above, at least one of the radicals R₁ and R₂ being other than hydrogen;

in the compounds of the formula (IIb) T is hydrogen or alkyl having 1 to 6 carbon atoms, T₁ is hydrogen, chlorine or alkyl or alkoxy having in each case 1 to 4 carbon atoms, n is 1 or 2 and, if n is 1, T₂ is chlorine or a radical of the formula -OT₃ or



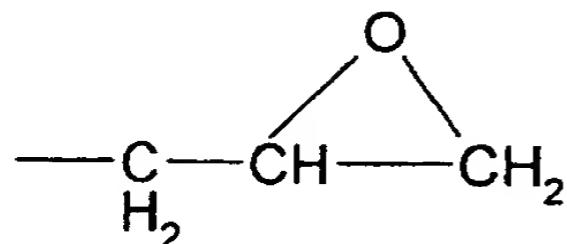
and, if n is 2, T₂ is a radical of the formula



or -O-T₉-O-

in which T₃ is hydrogen, alkyl which has 1 to 18 carbon atoms and is unsubstituted or substituted by 1 to 3 hydroxyl groups or by -OCOT₆, alkyl which has 3 to 18 carbon atoms, is interrupted once or several times by -O- or -NT₆- and is unsubstituted or substituted by hydroxyl or -OCOT₆, cycloalkyl which has 5 to 12 carbon atoms and is unsubstituted or substituted by hydroxyl and/or alkyl having 1 to 4 carbon atoms, alkenyl which has 2 to 18 carbon atoms and is unsubstituted or substituted by hydroxyl, phenylalkyl having 1 to 4

carbon atoms in the alkyl moiety, or a radical of the formula -CH₂CH(OH)-T₇ or



, T₄ and T₅ independently of one another are hydrogen, alkyl

having 1 to 18 carbon atoms, alkyl which has 3 to 18 carbon atoms and is interrupted once or several times by -O- or -NT₆-, cycloalkyl having 5 to 12 carbon atoms, phenyl, phenyl which is substituted by alkyl having 1 to 4 carbon atoms, alkenyl having 3 to 8 carbon atoms, phenylalkyl having 1 to 4 carbon atoms in the alkyl moiety or hydroxyalkyl having 2 to 4 carbon atoms, T₆ is hydrogen, alkyl having 1 to 18 carbon atoms, cycloalkyl having 5 to 12 carbon atoms, alkenyl having 3 to 8 carbon atoms, phenyl, phenyl which is substituted by alkyl having 1 to 4 carbon atoms, phenylalkyl having 1 to 4 carbon atoms in the alkyl moiety, T₇ is hydrogen, alkyl having 1 to 18 carbon atoms, phenyl which is unsubstituted or substituted by hydroxyl, phenylalkyl having 1 to 4 carbon atoms in the alkyl moiety, or -CH₂OT₈, T₈ is alkyl having 1 to 18 carbon atoms, alkenyl having 3 to 8 carbon atoms, cycloalkyl having 5 to 10 carbon atoms, phenyl, phenyl which is substituted by alkyl having 1 to 4 carbon atoms, or phenylalkyl having 1 to 4 carbon atoms in the alkyl moiety, T₉ is alkylene having 2 to 8 carbon atoms, alkenylene having 4 to 8 carbon atoms, alkynylene having 4 carbon atoms, cyclohexylene, alkylene which has 2 to 8 carbon atoms and is interrupted once or several times by -O-, or a radical of the formula -CH₂CH(OH)CH₂OT₁₁OCH₂CH(OH)CH₂- or -CH₂-C(CH₂OH)₂-CH₂-, T₁₀ is alkylene which has 2 to 20 carbon atoms and can be interrupted once or several times by -O-, or cyclohexylene, T₁₁ is alkylene having 2 to 8 carbon atoms, alkylene which has 2 to 18 carbon atoms and is interrupted once or several times by -O-, 1,3-cyclohexylene, 1,4-cyclohexylene, 1,3-phenylene or 1,4-phenylene, or T₁₀ and T₆, together with the two nitrogen atoms, are a piperazine ring;

in the compounds of formula (IIc) R'₂ is C₁-C₁₂alkyl and k is a number from 1 to 4;

in the compounds of the formula (III) u is 1 or 2 and r is an integer from 1 to 3, the substituents Y₁ independently of one another are hydrogen, hydroxyl, halogenomethyl, alkyl having 1 to 12 carbon atoms, alkoxy having 1 to 18 carbon atoms, or halogen, if u is 1, Y₂ is alkyl having 1 to 18 carbon atoms, phenoxy which is unsubstituted or substituted by hydroxyl, alkoxy having 1 to 18 carbon atoms, or halogen, or is substituted by alkyl or alkoxy

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having in each case 1 to 18 carbon atoms or halogen, alkyl which has 1 to 12 carbon atoms and is substituted by -COOH, -COOY₈, -CONH₂, -CONHY₉, -CONY₉Y₁₀, -NH₂, -NHY₉, -NY₉Y₁₀, -NHCOY₁₁, -CN and/or -OCOY₁₁, alkyl which has 4 to 20 carbon atoms, is interrupted by one or more oxygen atoms and is unsubstituted or substituted by hydroxyl or alkoxy having 1 to 12 carbon atoms, alkenyl having 3 to 6 carbon atoms, glycidyl, cyclohexyl which is unsubstituted or substituted by hydroxyl, alkyl having 1 to 4 carbon atoms and/or -OCOY₁₁, phenylalkyl which has 1 to 5 carbon atoms in the alkyl moiety and is unsubstituted or substituted by hydroxyl, chlorine and/or methyl, -COY₁₂ or -SO₂Y₁₃, or, if u is 2, Y₂ is alkylene having 2 to 16 carbon atoms, alkenylene having 4 to 12 carbon atoms, xylylene, alkylene which has 3 to 20 carbon atoms, is interrupted by one or more -O- atoms and/or is substituted by hydroxyl, -CH₂CH(OH)CH₂-O-Y₁₅-OCH₂CH(OH)CH₂, -CO-Y₁₆-CO-, -CO-NH-Y₁₇-NH-CO- or -(CH₂)_m-CO₂-Y₁₈-OCO-(CH₂)_m, in which m is 1, 2 or 3, Y₈ is alkyl having 1 to 18 carbon atoms, alkenyl having 3 to 18 carbon atoms, alkyl which has 3 to 20 carbon atoms, is interrupted by one or more oxygen or sulfur atoms or -NT₆- and/or is substituted by hydroxyl, alkyl which has 1 to 4 carbon atoms and is substituted by -P(O)(OY₁₄)₂, -NY₉Y₁₀ or -OCOY₁₁ and/or hydroxyl, alkenyl having 3 to 18 carbon atoms, glycidyl, or phenylalkyl having 1 to 5 carbon atoms in the alkyl moiety, Y₉ and Y₁₀ independently of one another are alkyl having 1 to 12 carbon atoms, alkoxyalkyl having 3 to 12 carbon atoms, dialkylaminoalkyl having 4 to 16 carbon atoms or cyclohexyl having 5 to 12 carbon atoms, or Y₉ and Y₁₀ together are alkylene, oxaalkylene or azaalkylene having in each case 3 to 9 carbon atoms, Y₁₁ is alkyl having 1 to 18 carbon atoms, alkenyl having 2 to 18 carbon atoms or phenyl, Y₁₂ is alkyl having 1 to 18 carbon atoms, alkenyl having 2 to 18 carbon atoms, phenyl, alkoxy having 1 to 12 carbon atoms, phenoxy, alkylamino having 1 to 12 carbon atoms or phenylamino, Y₁₃ is alkyl having 1 to 18 carbon atoms, phenyl or alkylphenyl having 1 to 8 carbon atoms in the alkyl radical, Y₁₄ is alkyl having 1 to 12 carbon atoms or phenyl, Y₁₅ is alkylene having 2 to 10 carbon atoms, phenylene or a group -phenylene-M-phenylene- in which M is -O-, -S-, -SO₂-, -CH₂- or -C(CH₃)₂-, Y₁₆ is alkylene, oxaalkylene or thiaalkylene having in each case 2 to 10 carbon atoms, phenylene or alkenylene having 2 to 6 carbon atoms, Y₁₇ is alkylene having 2 to 10 carbon atoms, phenylene or alkylphenylene having 1 to 11 carbon atoms in the alkyl moiety, and Y₁₈ is alkylene having 2 to 10 carbon atoms or alkylene which has 4 to 20 carbon atoms and is interrupted once or several times by oxygen;

in the compounds of the formula (IV) x is an integer from 1 to 3 and the substituents L independently of one another are hydrogen, alkyl, alkoxy or alkylthio having in each case 1

to 22 carbon atoms, phenoxy or phenylthio.

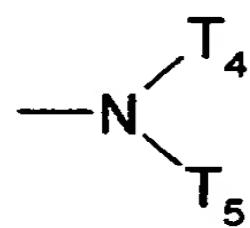
4. A polyolefin composition according to claim 3, wherein the substituent R₁ or R₂ in the compounds of the formula (IIa) is located in the ortho-position or para-position relative to the hydroxyl group.

5. A polyolefin composition according to claim 3, in which, in the compounds of the formula (IIa), R₁ is hydrogen or alkyl having 1 to 20 carbon atoms, R₂ is hydrogen, alkyl having 1 to 18 carbon atoms or phenylalkyl having 1 to 4 carbon atoms in the alkyl moiety and R₃ is hydrogen, chlorine or alkyl having 1 to 4 carbon atoms.

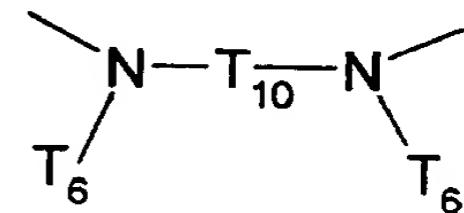
6. A polyolefin composition according to claim 5, in which R₁ is in the ortho-position relative to the hydroxyl group and is hydrogen or alkyl having 4 to 12 carbon atoms, R₂ is in the para-position relative to the hydroxyl group and is alkyl having 1 to 6 carbon atoms or cumyl and R₃ is hydrogen or chlorine.

7. A polyolefin composition according to claim 6, in which R₁ is alkyl having 8 to 12 carbon atoms, R₂ is alkyl having 4 to 6 carbon atoms and R₃ is hydrogen.

8. A polyolefin composition according to claim 3, in which, in the compounds of the formula (IIb), T is alkyl having 1 to 6 carbon atoms, T₁ is hydrogen, chlorine or alkyl having 1 to 4 carbon atoms, n is 1 or 2 and, if n is 1, T₂ is one of the radicals of the formula -OT₃ or



and, if n is 2, T₂ is a radical of the formula -O-T₉-O- or



in which T₃ is hydrogen, alkyl having 1 to 18 carbon atoms or alkyl which has 3 to 18 carbon atoms and is interrupted once or several times by -O-, T₄ and T₅ independently of one another are hydrogen, alkyl having 1 to 18 carbon atoms, alkenyl having 3 to 8 carbon atoms or hydroxyalkyl having 2 to 4 carbon atoms, T₆ is hydrogen or alkyl having 1 to 6 carbon atoms, and T₉ and T₁₀ are alkylene having 2 to 8 carbon atoms, alkenylene having 4 to 8 carbon atoms or alkylene which has 2 to 18 carbon atoms and is interrupted once or several times by -O-.

9. A polyolefin composition according to claim 8, in which T is alkyl having 1 to 4 carbon atoms, T₁ is hydrogen or chlorine and T₂ is one of the radicals of the formula -OT₃ or -O-T₉-O- in which T₃ is hydrogen, alkyl having 1 to 12 carbon atoms or a radical of the formula

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$-(OC_2H_4)_{4-8}-H$ and T_9 is alkylene having 2 to 8 carbon atoms.

10. A polyolefin composition according to claim 9, in which T_2 is a radical of the formula $-OT_3$ and T_3 is alkyl having 1 to 12 carbon atoms or a radical of the formula $-(OC_2H_4)_{4-8}-H$.

11. A polyolefin composition according to claim 3, in which, in the compounds of the formula (III), the substituents Y_1 are hydrogen, alkyl having 1 to 12 carbon atoms or halogen, if u is 1, Y_2 is alkyl having 1 to 18 carbon atoms, alkyl which has 1 to 12 carbon atoms and is substituted by hydroxyl, alkoxy having 1 to 18 carbon atoms, $-COOY_8$, $-CONY_9Y_{10}$ and/or $-OCOY_{11}$, glycidyl or phenylalkyl having 1 to 4 carbon atoms in the alkyl moiety, or, if u is 2, Y_2 is alkylene having 2 to 16 carbon atoms, alkenylene having 4 to 12 carbon atoms, xylylene or alkylene which has 3 to 20 carbon atoms, is interrupted by one or more $-O-$ atoms and/or is substituted by hydroxyl, the substituents Y_8 to Y_{11} being as defined in claim 3.

12. A polyolefin composition according to claim 11, in which the substituents Y_1 are hydrogen, alkyl having 1 to 4 carbon atoms or chlorine, if u is 1, Y_2 is alkyl which has 1 to 4 carbon atoms and is unsubstituted or substituted by hydroxyl, alkoxy having 1 to 18 carbon atoms, $-COOY_8$, $-CONY_9Y_{10}$ and/or $-OCOY_{11}$, glycidyl or benzyl, or, if u is 2, Y_2 is alkylene having 6 to 12 carbon atoms, butenylene, xylylene or alkylene which has 3 to 20 carbon atoms, is interrupted by one or more $-O-$ atoms and/or is substituted by hydroxyl, Y_8 being alkyl having 4 to 12 carbon atoms, alkenylene having 12 to 18 carbon atoms or alkyl which has 6 to 20 carbon atoms, is interrupted by one or more $-O-$ atoms and/or is substituted by hydroxyl, Y_9 and Y_{10} independently of one another being alkyl having 4 to 8 carbon atoms and Y_{11} being alkyl having 1 to 8 carbon atoms or alkenyl having 2 or 3 carbon atoms.

13. A polyolefin composition according to claim 12, in which u is 1 and r is 2, Y_1 is alkyl having 1 to 4 carbon atoms and Y_2 is alkyl having 1 to 18 carbon atoms or alkyl which has 1 to 12 carbon atoms and is substituted by hydroxyl, alkoxy having 1 to 18 carbon atoms, $-COOY_8$ and/or $-OCOY_{11}$, Y_8 being alkyl having 1 to 18 carbon atoms, alkenyl having 3 to 18 carbon atoms or alkyl which has 3 to 20 carbon atoms, is interrupted by one or more oxygen atoms and/or is substituted by hydroxyl, and Y_{11} being alkenyl having 2 to 18 carbon atoms.

14. A polyolefin composition according to claim 13, in which Y_2 is alkyl having 1 to 8 carbon

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atoms or alkyl which has 1 to 4 carbon atoms and is substituted by hydroxyl, alkoxy having 12 to 15 carbon atoms, -COOY₈ and/or -OCOY₁₁, Y₈ being alkyl having 8 to 12 carbon atoms, alkenyl having 12 to 18 carbon atoms or alkyl which has 5 to 10 carbon atoms, is interrupted by an oxygen atom and is substituted by hydroxyl, and Y₁₁ being alkenyl having 2 to 8 carbon atoms.

15. A polyolefin composition according to claim 13, in which Y₁ is methyl and Y₂ is an octyl radical or alkyl which has 1 to 3 carbon atoms and is substituted by hydroxyl, alkoxy having 13 or 15 carbon atoms, -COOY₈ and/or -OCOY₁₁, Y₈ being a decyl or octadecenyl radical or alkyl which has 7 carbon atoms and is substituted by hydroxyl and interrupted by an oxygen atom, and Y₁₁ being propenyl.

16. A polyolefin composition according to claim 3, in which, in the compounds of the formula (I), v and w independently of one another are 1 or 2 and the substituents Z independently of one another are hydrogen, halogen or alkoxy having 1 to 12 carbon atoms.

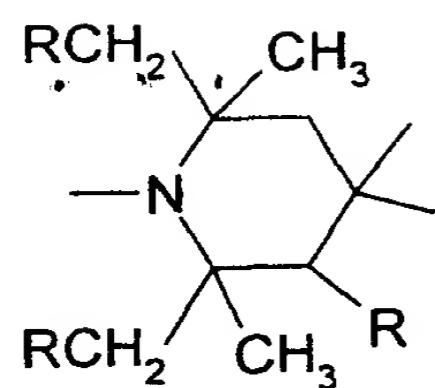
17. A polyolefin composition according to claim 3, in which, in the compounds of the formula (IV), x and y are 1 or 2 and the substituents L independently of one another are hydrogen or alkyl having in each case 1 to 12 carbon atoms.

18. A polyolefin composition according to claim 1 wherein the amount of the individual UV absorber in the mixture is from 20% to 80% based on the weight of the mixture, with the proviso that the sum adds to 100%.

19. A polyolefin composition according to claim 1 wherein the total amount of UV-absorber is from 0.005 to 5% based on the weight of the polymer.

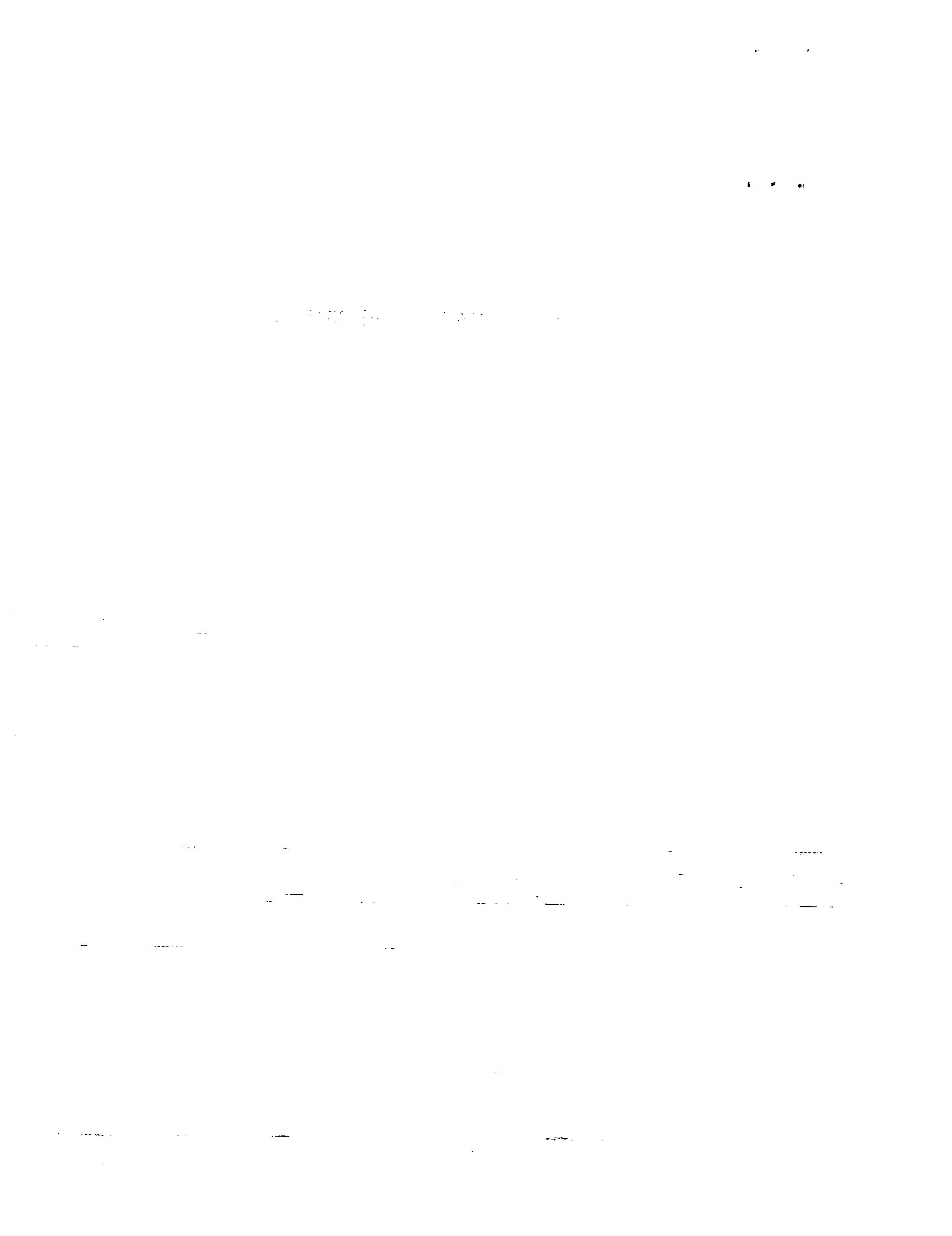
20. A polyolefin composition according to claim 1, which additionally contains at least one sterically hindered amine, in particular an amine of this type containing at least one radical of the formula

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in which R is hydrogen or methyl.

21. The use of a mixture of UV-absorbers according to claim 1 for the stabilization of polyolefins.



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Abstract of the invention

The present invention relates to polyolefin compositions which comprise as UV absorber a synergistic mixture of

- a) at least one hydroxybenzophenone and at least one 2-hydroxyphenylbenzotriazole with the proviso that the polyolefin is a high density polyethylene or a polypropylene of the "Phillips" type or a polyethylene or polypropylene of the metallocene type and no polyvinylpyridin is present;
- b) at least one hydroxybenzophenone and at least one 2-hydroxyphenyltriazine, with the proviso that if the polyolefin is polypropylene, no polyvinylpyridin is present
- c) at least one hydroxybenzophenone and at least one oxanilide;
- d) at least one 2-hydroxyphenylbenzotriazole and at least one oxanilide;
- e) at least one 2-hydroxyphenyltriazine and at least one oxanilide;
- f) at least one hydroxybenzophenone, at least one 2-hydroxyphenylbenzotriazole and at least one oxanilide;
- g) at least one hydroxybenzophenone, at least one 2-hydroxyphenylbenzotriazole and at least one 2-hydroxyphenyltriazine, with the proviso that if the polyolefin is polypropylene, no polyvinylpyridin is present;
- h) at least one hydroxybenzophenone, at least one oxanilide and at least one 2-hydroxyphenyltriazine; or
- i) at least one 2-hydroxyphenylbenzotriazole, at least one oxanilide and at least one 2-hydroxyphenyltriazine.

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